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On Kakishibu, IV.
Constitution of Shibuol, III.

By

Shigeru Komatsu and Naohiko Matsunami

(Received Feb. 8, 1928)

1. Tinctorial Properties of Shibuol and its Azoderivatives.

It was shown, in the first communication on this subject, that shibuol $C_{14}H_{20}O_9$ on fusion with caustic potash, gave phloroglucinol and gallic acid as a principal reaction product[1].

The natural colouring matters such as myricetin and delphinidin give equal rise by alkali fusion to the identical decomposition units as shibuol. These colouring matters and the tannin-like substance, however, show quite different properties from each other, in spite of being built up of the same constituents, due to the differentiation of the chemical structure of the complex nuclei in their molecules or of the manner of the linking up of the two constituents.

The facts which were pointed out by Perkin[2] that gallotannin and myricetin, a catechol tannin and quercetin, cyanomaclurin and morin, and butin and butein respectively occurring together as glucosides, or in a free state in flowers and in bark, lead us to suppose that, though they possess complex nature in composition, a certain resemblance exists between these colouring matters, as regards the similarity of their decomposition products, and there is reason to suppose that they may be structurally related.

The tinctorial property of the natural colouring matters, flavons,

1 These Memoirs A. 7. 15 (1923)

2 A. G. Perkin and A. E. Everest. The natural organic colouring matters (1918) P. 17.

flavonols, and anthocyans, and their azo-derivatives, according to the investigations by Buchner, Elsner and Kopp,[1] Willstaetter and Mallison[2] and Watson[3] affords some evidence for the determination of the number of the free hydroxyl group in the phenyl residue of the molecule of the colouring matters.

In order to acquire some evidence for the constitution of shibuol, the tinctorial property of the compound was investigated with that of myricetin, quercetin and catechin, in comparison with their metallic salts and benzene-azo-derivatives.

Benzene-azo-Shibuol.

8.1 grm. of fresh shibu free from pectin, and containing 5.5 grm. of shibuol, dissolved in sodium carbonate solution (14.5 grm. of the carbonate in 1540 cc. of water) were mixed with a diazobenzene chloride solution (3.7 grm. of aniline diazotized at 0° with 2.8 grm. of sodium nitrite in 100 c.c. of cold water and 9.8 grm. of hydrochloric acid (1.19) in 40 c.c. of water) at 0°–5°. After the coupling of the shibuol with the diazo-compound was completed, the temperature of the solution 5–15° was kept for 6 hours and then raised gradually to 50°, and the brownish yellow disazo-compound was filtered, washed with hot water and dried on a plate. The yield was 8 grm. (85% of the theory)

It was a reddish brown powder, fairly soluble in boiling alcohol and in nitrobenzene, but insoluble in ether and chloroform.

On analysis C=61.20; 61.65; H=4.27; 4.40; N=11.02; theory for $C_{14}H_{10}O_7$ $(C_6H_5N_2)_2$ C=62.40; H=4.00; N=11.2.

1 grm. of benzene-azo-shibuol dissolved in hot alcohol was reduced with a saturated solution of sodium hydrosulphite. After the solution had become colourless, it was acidified with dilute hydrochloric acid, and concentrated to small volume under reduced pressure, whereas a light-brownish substance was deposited, which was filtered and recrystallized from an alcohol solution, the yield was 0.5 grm. It was analysed after being dried at 100° under reduced pressure.

C=55.80; H=4.75; N=4.57:

theory required C=56.02; H=4.37; N=4.38 for $C_{15}H_{14}O_7N$.

The anlytical results of the reduced substance $C_{15}H_{14}O_7N$ indicated that one of the azo group in the benzene-azo-derivative, in the course of

1 A. G. Perkin & A. E. Everest: The natural organic colouring matters p. 257.

2 Liebig. Ann., **408**, 29 (1915).

3 J. Chem Soc., **107**, 1477 (1915).

reduction, was removed in the form of NH_3 from the molecule.

Benzene-Azo-Catechin.

The crude catechu prepared from gambier catechu (*Uncaria gambier, Roscough*) was used for the preparation of pure catechin[1].

The pure substance prepared according to the directions suggested by A.G. Perkin and E. Yoshitake[1] was in colourless needles, melting at 96° and on drying in a desiccator melting at 176°–177°.

Benzene-azo-catechin prepared by the method of Perkin and Yoshitake, and purified by recrystallization from a mixture of alcohol and nitrobenzene, was in salmon-red needles, melting at 194°–196°. The yield was 75% of the theory.

N=11.12 theory requires N=11,24 for $C_{27}H_{22}O_6N_4$

Benzene-Azo-Myricetin[2]

Myricetin which was prepared from the bark of the myrica rubra, was combined with diazo benzene chloride in the usual manner to form the benzene-azo-myricetin which, thus obtained was recrystallized from a mixture of alcohol and nitrobenzene. The yield was 83% of the theory.

It melts above 270°, and is soluble in nitrobenzene and in glacial acetic acid.

C=56.25; H=3.42; N=10.43; theory requires
C=56.60; H=3.10; N=10.64, for $C_{27}H_{18}O_8N_4$.

Benzene-Azo-Quercetin[3]

Quercetin from Merk was coupled with diazobenzene chloride in the usual manner. The yield was 70% of the theory. It was purified from the alcohol solution, and was found to melt at 235°–237°.

N=10.67; theory requires
N=10.98 for $C_{27}H_{18}O_7N_4$

In comparing the dyeing properties of these colouring matters, a quantity of shibuol or of flavones, corresponding to 5% by weight of the dyeing material-wools, was dissolved in a 5% acetic acid solution. Into this bath wools were dipped and left to soak for 50 minutes at a temperature of about 80°C. and the shades on the wools were shown

1 A. G. Perkin & E. Yoshitake; J. Chem. Soc., **81**, 1160 (1902). and also refer Clauser: Ber. D. Chem. Ges., **36**, 101 (1903),

2 S. Sato: J. Chem, Ind., (Japan) **17**, (1914)

3 A. G. perkin and Allison. J. Chem. Soc., **81**, 471 (1902).

in the following table:

Table I

Substance	Shade	Shade on wool after coupling with diazobenzene chloride
Shibuol	reddish brown	light brown
Catechin	yellow	pure orange
Myricetin	brownish yellow	brownish yellow
Quercetin	yellow	yellow

The shade given by shibuol was slightly darker than that by myricetin, but differs considerably from that given by catechin or quercetin.

Employing mordanted wool with chromium, aluminium, tin and iron, the following distinction of shade was observed, and these may be

Table II.

Substance	Chromium	Aluminium	Tin	Iron
Shibuol	yellowish brown	pale pink	pale violet	B'uish violet
Catechin	dark olive brown	〃 orange	—	yellow
Myricetin	reddish brown	brown orange	reddish orange	olive black
Quercetin	〃 〃	〃 〃	orange	〃 〃

due to different combinations of the two phenolicnucleus in the molecules of shibuol, catechin and myricetin.

Wool was dipped in a bath in which 5% of a benzene-azo-derivative of the above colouring substances was dissolved in 50% alcohol solution, and then acidified with 5% acetic acid. The shades on the wools were compared and the results are in the following table:

Table III.

Substance	shade	after treatment with iron mordant
Benzene-azo-shibuol	brown	greenish brown
〃-catechin	orange	orange 〃
〃-myricetin	yellowish brown	yellowish brown
〃-quercetin	brownish orange	brownish yellow

In dyeing property, shibuol and benzene-azo-shibuol greatly resemble myricetin and its benzene-azo-compound, but differ from catechin and quercetin and their azo derivatives in this respect. It was thus probable that the number of the free hydroxyl group in the phenyl-residue of a shibuol molecule closely resembles that of myricetion.

Definite evidence, however, of the constitution of shibuol is still lacking.

2. Reduction of shibuol.

The reduction of shibuol was, attempted in the following manner:

10 grm. of shibuol was dissolved in 200 c.c. of a 15% caustic soda solution and treated with 100 grm. of zinc dust for 3 hours at a temperature of 50°, the solution being acidified with sulphuric acid and the brown precipitate was separated, filtered and the solution evaporated to dryness under reduced pressure. The residue was extracted with alcohol and from the solution 0.1 grm. of gallic acid (m.p. 238°–234°) and 0.5 grm. of phloroglucinol (m.p. 210°) were isolated. The reduction of shibuol in an acetic acid solution with zinc dust at 50°, was tried, the result however, was a failure, no reduced substance being produced.

By the action of magnesium powder and acetic acid, shibuol was converted into a reddish-coloured substance soluble in the solution, and the behavior of shibuol was similar to flavones in this respect.

When, however, tetramethylshibuol was reduced with hydroiodic acid (1.18) at 130°, a violet brown powder was obtained as a reaction product, which is easily soluble in alcohol, acetic ether and acetone, but not soluble in ether. It resembled in composition a substance resulting from kakigoma by potash fusion, giving, on analysis, C=63.28% and H=3.97%.

Nov. 1924. Laboratory of Organic and Biochemistry.

On Kakishibu, V.
Methylation of Shibuol.

By

Shigeru Komatsu, Naohiko Matsunami and Masao Kurata.

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In the third report[1] on this subject, the writers reported on the acetylation of shibuol in order to find the number of free hydroxyl groups in its molecule, by which to ascertain the manner of combination between the two molecules of phloroglucinol and gallic acid, and it was supposed that shibuol contained at least four hydroxyl groups in the molecule, tetracetyl shibuol being the result.

Methylation of shibuol by means of dimethyl sulphate for the same purpose as that of the acetylation was undertaken, However, the process of methylation on flavones and tannins could not be employed in our case. In the methylation of shibuol, the reaction should be conducted at a low temperature such as 10°, to avoid decomposition of the shibuol by hydrolysis.

1. METHYLATION OF SHIBUOL IN A SODIUM CARBONATE SOLUTION.

100 c.c. of purified shibu consaining 10% of shibuol were mixed with a solution of 115 grm. of sodium carbonate dissolved in 150 c.c, of water, and to the mixed solution a great excess of dimethyl sulphate was added gradually with constant stirring during an interval of two hours, and the reaction was completed by stirring briskly at ordinary temperature for three hours, and then the temperature raised to 80°, and the reaction

1 These Memoirs, A, **8**, 235 (1925).

product separated, on cooling, filtered, washed with water, alcohol, and then ether, dried on a vacuum desiccator. The yield was 10.5 grm.

This was a violet brown powder, insoluble in alcohol, acetic ester, acetone, and ether. The methoxyl-value was estimated to be 17.0%, which corresponds to the value of dimethyl shibuol.

II. METHYLATION OF SHIBUOL IN CAUSTIC SODA SOLUTION.

100 c.c. of a 10% kakishibu solution were mixed with an alkaline solution of 46 grm. of caustic soda dissolved in 50 c.c. of water, at a temperature of 5°. 100 grm. of dimethyl sulphate were added during an interval of one hour to the mixed solution above mentioned, which was cooled with ice cold water after the reaction was completed, the temperature of the reacting product was raised gradually to 60°, and kept there for 30 minutes, and then the solution was left to stand at ordinary temperature over night. The yellowish brown precipitate thus formed, was filtered, washed with cold water and dried on a porous plate and on a vacuum desiccator. The yield was 12 grm. It began to sinter at 220° and melted at 260° with decomposition.

It was soluble in alcohol, benzene, chloroform, acetic ester, and acetone, and also in dilute caustic alkali solution, but insoluble in water, and in ether. The analytical results agree well with these of trimethyl shibuol.

C=60.78 ; 60.83 ; H=5.31 ; 5.42 ; OCH_3=27.95.

C=61.07 ; H=5.38 ; OCH_3=27.84, Calc. for $C_{11}H_9O_4(OCH_3)_3$.

III. METHYLATION OF SHIBUOL IN DILUTE CAUSTIC SODA SOLUTION AT HIGH TEMPERATURE.

10 grm. of purified shibuol were dissolved in 325 grm. of a 20% caustic soda solution at 20°, to the mixture 100 grm. of dimethyl sulphate were added during an interval of 30 minutes, and the mixture left for one hour. The reaction mixture was then heated to 30° on a water bath, and a violent reaction was observed to take place instantly. After the reaction had been completed, the product was boiled for 30 minutes, and the precipitate (A) formed, separated by filtration from the mother liquor, washed with water, and treated with alcohol.

The product (A) was composed of two substances, one was insoluble in alcohol, and the other soluble in alcohol, acetone, and acetic ester; the former was a brown powder, sintering at 200°, and melting at 230°-240° ; and the latter substance was a pale yellow amorphous powder.

The mother liquor separated from the substance (A), on acidifying the solution with mineral acid, yielded a brown precipitate (B) which was purified from the alcohol solution, and was soluble in acetone, acetic ester, and acetic acid.

On analysis of the substances (A) and (B), the following results were obtained:

A. C=63.48; 63.44; H=5.40; 5.60; OCH_3=34.8.

B. C=61.70; 61.22; H=4.49; 4.82; OCH_3=23.5; Ash=0.2,

The substance (A) was determined to be tetramethyl shibuol, and the (B), trimethyl shibuol.

IV. METHYLATION OF SHIBUOL IN AN ACIDIC MEDIUM.

When shibuol was methylated with dimethyl sulphate in an alkaline medium, dimethyl and trimethyl shibuol resulted as reaction products; but the reaction which was carried out at high temperature, proceeded to form mostly di-or tri-methyl shibuols with some tetramethyl shibuol.

The writers, however, applying the methylation with dimethyl sulphate in an acidic medium, have succeeded in obtaining tetramethyl shibuol with fruitful results:

1 Mol of purified shibuol dissolved in a 10% caustic soda solution (5 mol of NaOH in water), and to the solution 12 mols. of dimethyl sulphate were added under cooling with ice-cold water with constant stirring. After standing the reaction mixture for 3-4 hours, 15 mols. of caustic soda in a concentrated solution were added and the mixture was left to stand at room temperature for one day. Tetramethyl shibuol, by this treatment, was separated in a reddish brick-coloured precipitate from the solution, and this was filtered and washed with water. The yield of the crude substance was 55 grm. from 50 grm. of shibuol, which corresponds to 92% of the theory.

It shrinks at 200° and melts at 220°. C=56.49; H=5.04; OCH_3=34.44; ash=3.9.

The crude tetramethyl shibuol was divided into two parts by their solubility in an acetone-alcohol solution, The crude substance was dissolved in cold acetone, and alcohol was added, whereupon the precipitate (C), was formed, separated from the solution by filtration, the solvent was then mainly expelled from the solution by distillation, and by adding alcohol to the residue, the second substance (D) was separated. Both substances were analysed:

	C	H	Ash	OCH_3
C.	60·19	5·77	2·06	34·85
	60·24	5·44	1·93	——
D.	62·48	6·42	0·1	38·10
	63·01	6.84	——	——

The tetramethyl derivative was observed as in the case of acetyl shibuol to be separated by means of a mixture of acetone and alcohol into the soluble and insoluble forms in the solvent. They show the same elementary composition and the methoxyl value, but they differ in solubility and ash-content from each other and such a relation of chemical & physical properties between the two substances is the same as that observed in shibuols and their acetyl compounds.

71 grm. of the crude tetramethyl shibuol by fractional precipitation of its acetone-alcohol solution, were divided into the following four fractions which show the melting points, and solubility in acetic ester, acetone, and alcohol.

	I	II	III	IV
Shrinks at	275°	260°	150°	90°
Melts at	288°*	270°	185°*	130°
Colour	light drown	light brown	reddish brick colour	yellowish brown
Acetone	soluble	soluble	soluble	soluble
Acetic ester	〃	〃	〃	〃
Alcohol (cold)	sparingly soluble	——	〃	〃
Ether	insoluble	——	sparingly soluble	somewhat soluble
Caustic soda	〃	——	〃	〃
Yield in grm.	31	7.5	11	8.5
Yield in %	53	13	19	15

*with decomposition.

V. METHYLATION OF TRIMETHYL SHIBUOL.

With 5 grm. of trimethyl shibuol dissolved in 30 c.c. of acetone, 10 grm. of methyl iodide, and 8 grm. of silver oxide were mixed, and the mixture was shaken in a sealed tube for 5 hours, and the reaction product separated from the silver iodide was purified from acetic ester. The yield was 6 grm.

It was a light brown powder and soluble in alcohol, acetone, and acetic ester, but insoluble in alkali solution, and melting at 208°–210° with decomposition. Analytical results agree with those for tetramethyl shibuol.

C=61·70; 61·89; H=5·70; 5·81; OCH_3=34·47; theory requires C=62·06; H=5·74; OCH_3=35·63 for $C_{11}H_8O_3$ $(OCH_3)_4$.

VI. ACETYLATION OF TRIMETHYL SHIBUOL.

Trimethyl shibuol was acetylated with acetic anhydride and sodium acetate in a sealed tube heated at 140° for two hours. The reaction product was purified with acetone and then its acetyl value determined. CH_3CO=9.77% agrees with the theoretical value 11.16% for monoacetyl trimethyl shibuol. It was soluble in acetone and in acetic ester, but insoluble in water and in alcohol.

Shibuol when methylated gave a tetramethyl compound, a fact which indicates the presence of at least four free hydroxyl groups in the molecule, and harmonizes with the experimental results obtained by the acetylation.

Oct. 1927, Laboratory of Organic- and Bio-Chemistry.

An Examination of the Micro-Crystals of Calcium Carbonate in Molluscan Shells by means of X-Rays. Part I.

By

Jinzo Tsutsumi

(Received Feb. 15, 1928)

Abstract

Examination of the micro-crystals in the prismatic layer of *Atrina japonica* REEVE by means of the X-rays convinced the writer that the micro-crystals in this layer are in the form of calcite, and that the crystals are arranged fibrously, the principal axes of the crystals being parallel to the direction perpendicular to the surface of the shell. This is in accordance with the results of observations made by different means by the previous authors.

The problems of whether the micro-crystals of calcium carbonate in molluscan shells are in the form of the calcite or in that of the aragonite and of what their arrangement is, have already been studied by G. Rose and others[1] physically, microscopically and chemically, but as such means are always not without defect, it appeared to me more or less hopeful to attack the same problems by means of x-rays.

Although the shells are not the same in chemical composition or physical structure in different species of animals, they are mainly composed of pure calcium carbonate, an organic base known as conchiolin, allied to chitin, and a small amount of a phosphate of calcium.

The shell of *Atrina japonica* REEVE or *Tairagi* as it is commonly called in Japanese, consists of three layers: a very thin periostracal or the uppermost layer, a prismatic or middle layer and, a nacreous or the lowest layer. The present investigation is concerned mainly with the prismatic layer. The prismatic layer shows a columnar structure

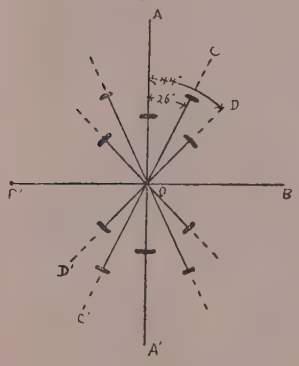
1 G. Rose, G. Bournon, De La Beche, Necker: Winterstein :- Handbuch der vergleichenden Physiologie, Bd. III, Teil 1, Heft 1.

and the axes of the columns are nearly parallel to the direction perpendicular to the surface of the shell. The carbonate of calcium in this layer is already known to be in the form of calcite and this has been confirmed by the writer too by means of Hull's powdered crystal method.

Then, in order to examine the arrangement of the micro-crystals of calcite in the prismatic layer the writer took the Laue-photographic method. The x-rays used for the device were from a molybdenum-target in a Coolidge-tube. A pair of narrow circular slits or apertures were used to get a narrow beam of x-rays, and a photographic plate was set perpendicularly to this beam of x-rays. A specimen of the prismatic layer, about 0.7 mm. in thickness, was put just behind the second slit which was against the photographic plate. The distance between the specimen and the photographic plate was 4.1 cm. throughout the experiments.

The photograph reproduced in Fig. 1, Plate I, is one obtained with the prismatic layer of the *Atrina japonica* Reeve, the axis of the columns having been set perpendicularly to the incident x-ray beam in this case. We see that the pattern in this picture consists of many intense spots about a central spot impressed by the direct beam of the x-rays and some radiating bands starting from the central spot and passing outward through the intense spots.

Fig. 1



The accompanying figure shows such a pattern diagrammatically. The line AOA′ has been drawn parallel to the axis of the column of the layer and COC′, DOD′ represent the successive bands arranged symmetrically about the vertical and horizontal lines AOA′, BOB′. There is no doubt that the radiating bands are due to the reflection of X-rays from various atomic planes of the micro-crystals which are arranged fibrously. Fig. 2, Plate I, shows the next case, in which the axis of the column of the layer was set parallel to the x-ray beam, and the appearance of concentric rings in this case, instead of the radiating bands as in the former case, indicates that the axis of the fibrous arrangement of micro-crystals coincides nearly at least with the axis of the column of the layer. I experimented in this way with the various portions of the prismatic layer of the shell, and

no difference was ever detected so far as the present experiment is concerned.

Since the spots on the radiating bands owe their origin to the reflection of the K–radiations of molybdenum from the various atomic planes of the crystals, we can find the indices of these atomic planes from the distances between these spots and the central one. Let us denote the distance between a spot and the central one by r, the distance between the photographic plate and the specimen by a and the glancing angle of the beam of x–rays to an atomic plane which is to be determined, by θ. Then the value of θ will be calculated by the following equation :

$$\tan 2\theta = \frac{r}{a}.$$

Finding the value of θ in this manner and by taking 7.1 A. U. as the mean wave–length of the K–lines of molybdenum, the spacing between the consecutive atomic planes will be obtained by Bragg's equation :

$$n\lambda = 2d \sin\theta$$

where n is an integer, and is taken to be unity in the present case. The spacings d corresponding to various radiating bands thus determined are tabulated in Table I, and are compared with those observed by W. L. Bragg[1]. As the two results are in good accordance with each other we can safely assign the indices given by Bragg to the corresponding bands.

Table I

Bands	a	d in A. U. (obs.)	d (Bragg)	Indices.
A O A′	0°	2.84	2.86	(111)
C O C′	26°20′	1.92	1.91	(110)
D O D′	44°10′	3.03	3.03	(100)

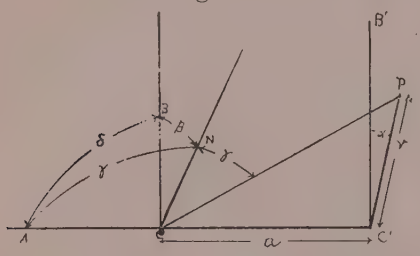
Thus the indices of the reflecting atomic planes responsible for every radiating band are known, while the crystallographic axis of the calcite, which is in the direction parallel to the axis of the fibre, will

1 Roy. Soc. Proc. London. Vol. **105**, (1924)

be known from the angular distribution of these radiating bands. The angles α between the lines connecting the central spot and the intense spots on every radiating band and the direction A O A′ on the photographic plate, which is parallel to the axis of the fibre, are also given in the second column of Table I.

A C C′ in Fig. 2 represents the beam of the incident x-rays which makes an angle δ with the direction of the axis of the fibre C B, and C N the normal to a reflecting atomic plane which makes the angles β and γ with C B and C A respectively. So the angle γ is the angle of incidence to the atomic plane under consideration, and is the complement of the glancing angle θ. The x-ray beam reflected from the atomic plane falls at P on the photographic plate B′ C′ P standing normally to the incident beam at C′, the line B′ C′ on the photographic plate being here drawn parallel to the line B C. Now, in the spherical triangle A B N we have the following relation:

Fig. 2



$$\cos\beta = \cos\gamma \cos\delta + \sin\gamma \sin\delta \cos\alpha,$$

where α is the angle between the plane B C A and the plane N C A. If A C is perpendicular to C B as in the case of Fig. 1, Plate I, the above relation can be reduced to $\cos\beta = \sin\gamma \cos\alpha$. Since γ is known for a given wave-length of x-rays and for a definite reflecting atomic plane, the values of β, the angle between the axis of the fibre and the normal of a reflecting atomic plane, can be readily obtained from the observed values of α respectively. The values of β thus determined are tabulated in Table II. It has already been stated

Table II.

Bands	Indices	β (obs.)	β (calc.)
COC′	(111)	26°40′	26°22′
DOD′	(100)	44°20′	44°40′

that the band ACA′ in Fig. 1 lies, nearly at least, parallel to the axis of the fibre; moreover it was shown in Table I that this band is caused by

the reflection of x-rays from the atomic plane 111. From these facts it seems natural to take the principal axis 111 of the calcite as the axis of the fibre, and the values of β calc. in the fourth column of Table II are calculated under this consideration. Such fair agreement between the observation and the calculation seems to show reversely that the principal axis of the calcite is the axis of the fibrous arrangement. This was also tested by using a spherical scale and a globe devised by U. Yoshida[1], and it was observed that the distribution of the radiating bands under consideration could not be accounted for in any way other than this. Here it must be noted that, if the principal axes of all the micro-crystals were arranged exactly in the same direction, the band corresponding to the plane 111 could not appear on the photograph; but as a weak impression of the band is detectable in the picture, we must consider that the parallelism of the principal axes of the micro-crystals is not very exact; and that, though the majority of the micro-crystals are arranged nearly in the ideal orientation, some of them deviate a little from that direction.

Thus the results of my experiment with x-rays are the same as those obtained by different means by the previous authors[2]: and it seems to me entirely established that the micro-crystals in the prismatic layer of the shell *Atrina japonica* REEVE are calcite and that the principal axes of these micro-crystals of calcite lie almost parallel to the direction of the column in this layer.

Lastly it must be noted that the micro-crystals of this shell are rather coarse, since the x-ray photographic picture appears as an assemblage of many distinct Laue-spots. It is this circumstance that hindered a little the accurate determination of the positions of the bands, though the conclusion is not to be disturbed. My examination of the arrangement of the micro-crystals of aragonite in the nacreous layer of the shell of the same animal is now going on and the results will be published in the near future.

In conclusion, I desire to express my sincere thanks to Professors U. Yoshida and T. Kawamura for their kind supervision throughout this research.

1 U.Yoshida; Jap. J. Phys., **4**, 133(1927)

2 G. Rose, V. Ebner, Leydolt, Valentin: Winterstein:- Handbuch der vergleichenden Phsyiologie, Bd. III, Teil 1, Heft 1.

Plate I

Fig. 1

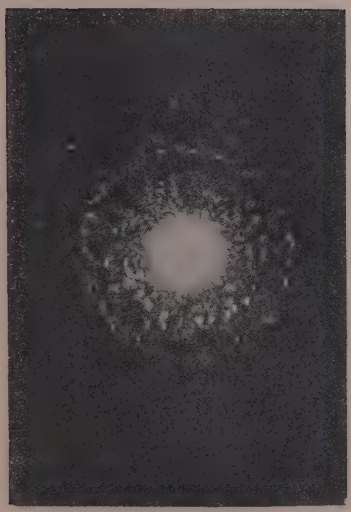
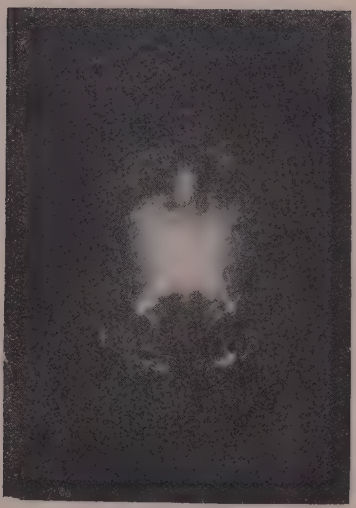


Fig. 2



The Arrangement of Micro-Crystals in the Banded "Supposed Aragonite (Arareishi)".

By

Hidehiko Fukushima

(Received Feb. 15, 1928)

Abstract

By means of x-rays it was ascertained that the banded "supposed aragonite" is an aggregate of micro-crystals of calcite, and that most of them are so arranged that their principal axes are perpendicular to the surfaces of the layers in the mineral.

Nature sometimes produces banded minerals. It is naturally supposed that, in these minerals, the micro-crystals are arranged in some regular manner. If so, the determination of their manner of arrangement in various banded minerals seems to be of much interest, as it seems to have some connection with the problem of the atomic forces. This has already been studied microscopically in the case of some minerals, but even this method is not always without defect, and it is not applicable to all cases. Thus the writer employed the x-rays, and examined the arrangement of the micro-crystals, first, in the "supposed aragonite (arareishi)," the banded mineral whose constituent was proved to be calcite and not aragonite, as will be described below.

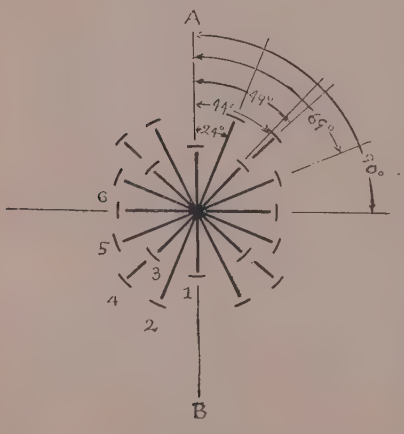
The specimen used by the writer was the "arareishi" as it is usually called by mineralogists, and which is produced at Oshikamura, Shinano, Japan. As shown in Fig. 1, Plate I, it is a banded block of light brown colour; and the writer confirmed, by the powder photograph method, that it is composed actually of calcite and not of aragonite. The "arareishi" obtained at Tahiramura, Shinano, which had been usually believed by mineralogists to be composed of aragonite, has

already been proved by means of x-rays to be calcite by Y. Ozawa[1], and he called that mineral "supposed aragonite." The writer will assign this name also to the "arareishi" produced at Oshikamura, Shinano.

At first, a specimen in the form of a thin plate of about 0.5 mm. in thickness was cut out along the layers of the supposed aragonite; and a narrow beam of x-rays, radiated from a Coolidge tube with a molybdenum target, was made to strike the specimen in the direction perpendicular to the surfaces of the layers of the specimen. The photographic plate was placed perpendicularly to the incident beam of x-rays at a distance of 3.5 cms. behind the specimen. The photograph thus taken is shown in Fig. 2, Plate I. The presence of concentric rings only in the photograph informs us that the orientation of the micro-crystals is symmetrical about the normal to the surfaces of the layers.

Next, the writer prepared another specimen by stripping it off perpendicular to the layers from the same block of arareishi as before. An x-ray beam was made to strike the specimen in a direction perpendicular to the normal of the surfaces of the layers, and the diffraction pattern was obtained on a photographic plate placed perpendicularly to the incident x-ray beam at a distance of 4.2 cms. behind the specimen. The diffraction pattern thus obtained is reproduced in Fig. 3, Plate I. As is seen in the figure, we can detect 20 prominent bands radiating outward from the central spot. The intense spots situated at the ends of these bands were confirmed to correspond to the K_α radiation reflected from various atomic planes of calcite.

Fig. 1



In Fig. 1. the distribution of the bands and spots in Fig. 3, Plate I are represented diagramatically. In this figure, the direction A B is drawn parallel to that of the normal to the surfaces of the layers in the specimen. As can be seen from this figure, or from Fig. 3, Plate I, the distribution of the bands and the spots is symmetrical about the line A B. This is in accordance with

1 Y. Ozawa : Science Reports, Tohoku, **14**, 33(1925)

the fact that the orientation of the micro-crystals is symmetrical about the direction of the normal to the surfaces of the layers; and consequently we may infer that the supposed aragonite has a fibrous structure whose axis is in the direction perpendicular to the surfaces of the layers of the specimen.

The next problem is to find out the crystallographic axis which is arranged in the direction parallel to the axis of the fiber. For this purpose the writer used the values of θ and β obtained from the position of the spots and the angular distribution of the bands[1]. Here θ is the glancing angle of the K_α radiation of molybdenum to an atomic plane of calcite, and β the angle between the normal to an atomic plane and the crystallographic axis of the fiber which is to be determined. The observed values of θ and β are tabulated in Table I. The indices

Table I

No. of the bands and the spots	Indices of the planes	Spacing d in A.U.	Distance from the centre to the spot(mm)	$\theta_{calc.}$	$\theta_{obs.}$	$\beta_{calc.}$	$\beta_{obs.}$
1	(111)	2.86	10	7°11′	7°	0°	0°
2	(110)	1.91	16.5	10°43′	11°	26°32′	26°
3	(100)	3.03	10	6°44′	7°	44°40′	44°
4	$(\bar{1}22)$	2.02*	16.5	10° 7′	11°	44°37′	49°
5	$(12\bar{2})$	2.30*	14	8°53′	8°	74°19′	69°
6	$(\bar{1}10)$	2.49	12.5	8°12′	8°	90°	90°

of the atomic plane which is responsible for a radiating band is obtained from the value of θ, and are given in the second column of Table I. From the values of β and the indices for every radiating band, and by using the globe and the spherical scale devised by U. Yoshida[2], the crystallographic axis of the fiber was found to be the principal axis 111 of the calcite crystal: i.e. the principal axes of the microcrystals of calcite in the supposed aragonite were found to be arranged parallel to the axis of the fibre which is perpendicular to the surfaces of the layers. This arrangement of the micro-crystals of calcite is the same as that observed by Mr. J. Tsutsumi[3] in the prismatic layer of a shell.

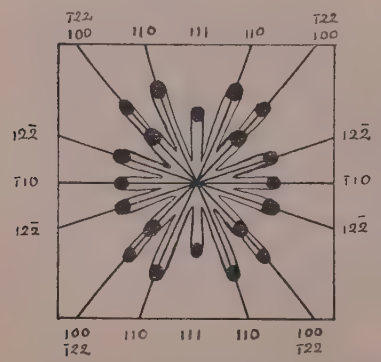
1 S. Nishikawa : Su. Butu. Kw. K., Tokyo, **7**, 131 (1913-1914)
2 U. Yoshida : Jap. Jour. of Phys, **4**, 133 (1927)
3 J. Tsutsumi : These Memoirs, A. **11**, (1928).

The calculated and the observed values of θ and β are compared in Table I, and the discrepancies between them seems to be within the limit of experimental errors. As to the planar spacings d, the values observed by W. L. Bragg[1] are given in the table for 4 different atomic planes, and for the two others they were calculated by using Bragg' data : $d_{100}=3.03$ A. U. and they are marked by asterisks in Table I.

If the principal axes of all the microcrystals are exactly parallel to the axis of the fibre, the spots 1, which seems to be caused by the plane (111), are not able to present themselves. Consequently it must be considered that, though the majority of the micro-crystals in the supposed aragonite are arranged in the fibrous structure before mentioned, there are some whose orientations deviate a little from the ideal one. The fact that the bands are not sharp but diffused, is well in accordance with this consideration. In order that the reflection of the K_α line of molybdenum may occur from the atomic plane (111), the glancing angle must be 7° 11′. Therefore the deviation of the principal axes of some of the micro-crystals from the ideal orientation must be at least 7° 11′. But at the same time, the absence of the concentric rings which usually appear besides the radial bands, in the case of Aluminium wire[2] or others, implies that scarcely any micro-crystals with much more deviation than the critical angle above mentioned, are present.

Being coloured light brown as stated before, the supposed aragonite seems to involve some impurities. But, as the spots and the bands in Fig. 3, Plate I are all explainable by the reflection of x-rays from calcite crystals only, the impurities, if any exist, seems to be of very small amount.

Fig. 2



The curves plotted in Fig. 2 represent the calculated ones obtained by taking the principal axis of the calcite crystal as the axis of the fibre, and by considering that the axis of the fibre and the photographic plate are both perpendicular to the incident x-rays. The positions of the bands and the spots on the photograph are shown in the figure. The agreement between the observation and the calculation

1 W. L. Bragg : Roy. Soc. Proc., **105**, (1924)
2 T. Fujiwara : Mem. Coll. Sci. Kyoto, A. **8**, (1925)

may be easily seen in this figure. The reason why the bands and the spots due to the atomic plane (111) appeared on the photograph, has already been stated; and the theoretical curve corresponding to this atomic plane is omitted in the figure.

In conclusion, the writers' hearty thanks are due to Prof. U. Yoshida for his kind guidance in this research, and the writer also expresses his sincere thanks to Mr. J. Tsutsumi for his various suggestions.

Plate I.

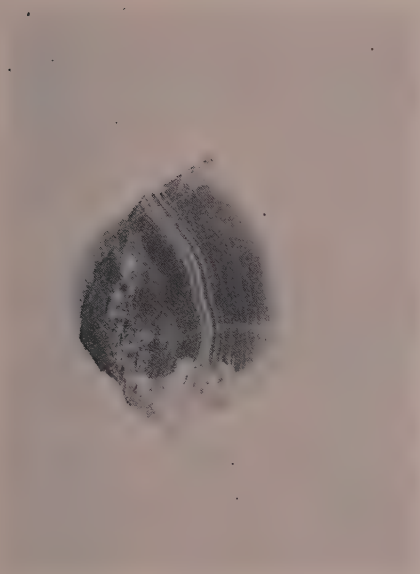


Fig. 1

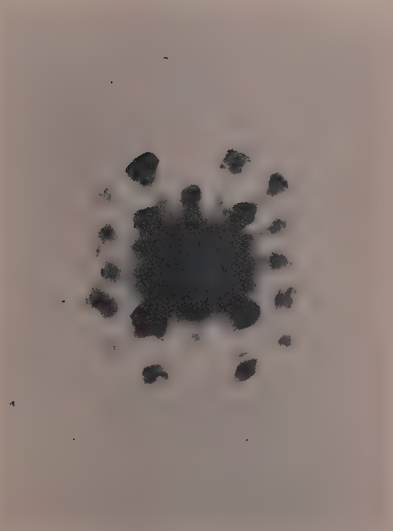


Fig. 3

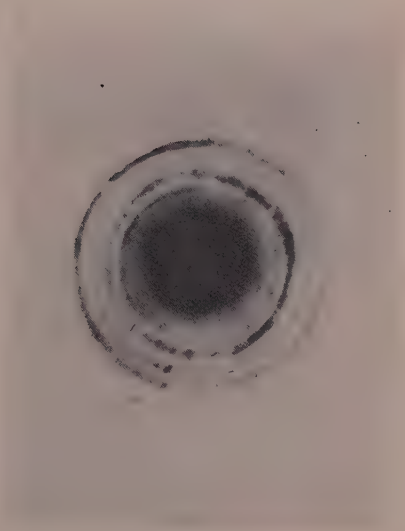


Fig. 2

The Recrystallization of Aluminium

By

Kenzo Tanaka

(Received Feb. 15, 1928)

Abstract

The orientations of the single crystals formed by recrystallization in the extended wires and plates of single crystals of aluminium were examined by means of X-rays, and the orientations were observed to be rather at random in both cases, irrespective of the grade of extension and the orientations of the initial crystals.

When a deformed metal is subjected to suitable heat-treatment, there grow crystals of new orientation in a specimen of a deformed single crystal as well as in one composed of an assemblage of micro-crystals. In a previous experiment,[1] the writer examined the orientations of single crystals of aluminium formed in specimens composed of micro-crystals.

In the present research, the writer has determined the orientations of 192 crystals of aluminium obtained by recrystallization from deformed single crystals, to see whether there is some relation between the orientations of crystals before and after the recrystallization or not. The orientation of the crystals was determind by treating the distribution of Laue-spots with the globe and the spherical scale devised by U. Yoshida.[2]

The specimens were taken from commercial aluminium wires of 3 mm. in diameter and plates of 1.5 mm. in thickness. To let the crystal grow, the wires were drawn through circular dies and the plates were

1 K. Tanaka, Jap. J. Phys., **4**, 137 (1927)
2 U. Yoshida, Jap. J. Phys., **4**, 133 (1927)

extended by 1 or 2 per cent before annealing. The orientations of the large crystals thus formed were determind before recrystallization. The distribution of the orientations of the crystals thus obtained showed the same feature as noted in the previous paper, and it was observed that, in the case of the wire, the [210] axis of the crystal was more favoured by the direction of the axis of the wire than any other crystallographic axis.

These specimens were then extended by various percentages by pulling, and annealed at about 600°C. for 5 hours to let them recrystallize. Fig. 1, Plate I shows the Laue-photograph of an undeformed aluminium single crystal wire taken by the method of using a long slit[1]. Figs. 2 and 3 Plate I and Fig. 4 Plate II were taken by the same method and show the changes in the orientation of the crystal and the internal deformation of the same specimen when it was extended by 2, 10 and 42 (break) per cent respectively by pulling. As can be seen in these figures, the diffraction lines change their positions and become diffuse and irregular as the degree of extension increases. This shows that the single crystal is broken, by the process of pulling, into a large number of smaller pieces with the orientations scattered to some extent; and that, at the same time, the average orientation of these smaller crystals is altered gradually.

It has already been observed by Karnop and Sachs[2], and Arkel and Bruggen[3], that when the extension is too small, recrystallization does not take place in the case of an aluminium single crystal. In the case of Al wires of from 1 to 3 mm. in diameter, this critical per cent of elongation for the recrystallization to be effective was observed by the writer to be about 3 per cent from the identity of the distorted Laue-spots before and after the annealing. On the other hand, when the extension was larger than about 20 per cent the recrystallized grains were too small for Laue-photographs of them to be taken separately. Therefore, the range of the extension investigated here was from 4 to 20 per cent, and the corresponding linear sizes of the recrystallized grains varied mostly from about 0.4 to 8 cm.

The orientations of the crystals obtained by recrystallization were rather random, and no particular relation was detected between the initial crystals and those obtained by recrystallization, irrespective of the

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1 U. Yoshida and K. Tanaka, Nature, **118**, 912 (1926)

2 R. Karnop and G. Sachs, ZS. f. Physik, **42**, 283 (1927)

3 A. E. V. Arkel and M. G. v. Bruggen, ZS. f. Physik, **42**, 795 (1927)

grade of extension applied to the specimens before recrystallization. This seems to be of no little interest; and it is especially worth noting that the crystals of new orientation come out and grow in a specimen of small extension, the orientation of small crystals formed by extension in this specimen being nearly the same as that of the original single crystal.

It has been stated that the orientations of the new crystals formed by recrystallization in the extended wires of single crystals are rather random. But this is only a general statement, and it would be more exact to say that the vicinity of the [111] axis of the crystal seems to be a little favoured by the direction of the axis of the wire, so far as the present experiment is concerned. Two examples of the orientations of the axes of two wires before and after the recrystallization, in refer-

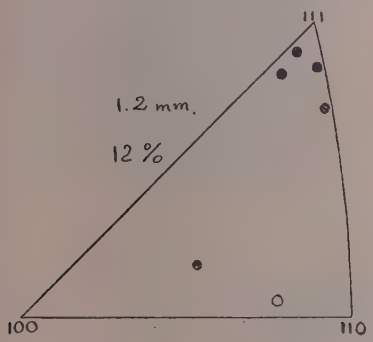


Fig. 1

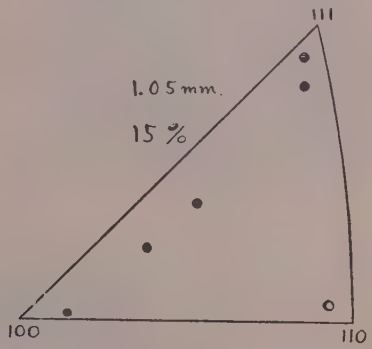


Fig. 2

ence to the crystallographic axes, are shown separately in Fig. 1 and 2, by the stereographic projection. Here 100, 110 and 111 represent the directions of the normals to the atomic planes (100), (110) and (111) respectively, and the small circle represents the direction of the axis of the wire before the recrystallization and the dots in the same figure those in separate pieces of single crystals obtained by recrystallization. The diameter of the wires and the percentage of the extension are written separately in each figure.

For the specimens in the form of plate, the lengthwise directions and the surface normals of them were represented in reference to the crystallographic axes, and it was observed that the orientation of the crystals formed by the recrystallization was rather at random, and no particular relation between the orientation of the crystals formed by recrystallization and that of the initial ones could be detected.

Four photographs in Fig. 5, Plate. II show the appearances of the crystal grains before and after the recrystallization when the specimens of single crystals were subjected to extensions of 4, 6, 8 and 10 per cent respectively before the annealing process. The boundaries of initial crystals are shown by the lines marked on the surfaces of the plates and those of the recrystallized grains will be easily detected from the difference in the shades of the photographic images at various parts of the surfaces of the specimens.

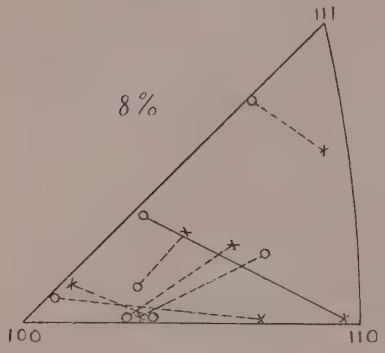


Fig. 3

In Fig. 3, for example, the direction of the surface normal and the lengthwise direction of a plate are shown, in reference to the crystallographic axes, by the stereographic projection as in the case of the wire. Here the direction of the surface normal and the lengthwise direction of the plate are indicated by a circle and a cross respectively, and the line connecting a circle and a cross indicates that they belong to the same crystal. Of the lines thus joining a circle and a cross, the full line means that the circle and the cross connected by this line belong to the crystal examined before recrystallization, and the remaining broken lines relate to the crystals obtained by the recrystallization at the place formerly occupied by the crystal signified by the full line.

In conclusion, the writer wishes to express his sincere thanks to Professor U. Yoshida for his kind guidance in the research.

Plate I

Fig. 1

Fig. 2

Fig. 3

Plate II

Fig. 4

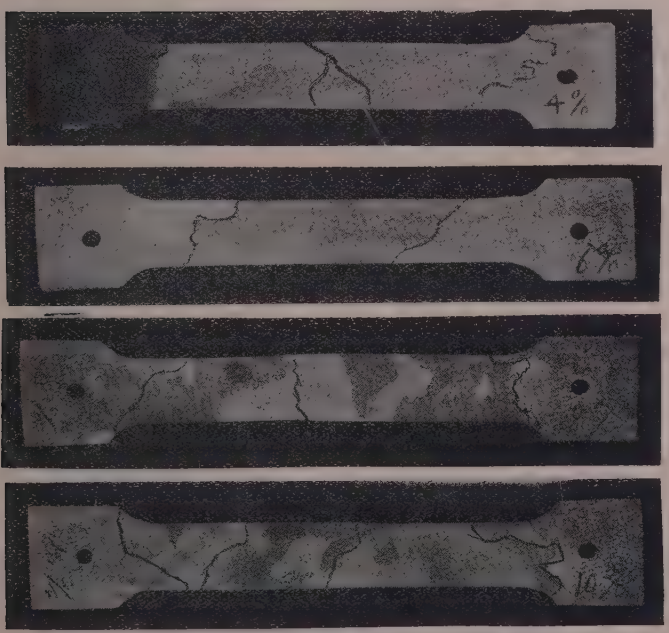


Fig. 5

The Influence of the Heliographic Activity upon the Solar Constant,[1] with Two Appendices.

By

Issei Yamamoto.

(Received April 12, 1928)

Abstract.

Abbot and his colleagues detected years ago some kinds of interrelations apparently existing between the variations of the solar constant and heliographic phenomena. A special attack on this problem has been made here from rich materials preserved in the Mount Wilson Observatory. Not only have the relations suggested by our forerunners been confirmed, but some similar cases extending the idea of these relations have been detected. Sudden drops of the solar heat corresponding to the central meridian passage of some solar activity area on the visible disc are seemingly due to the absorbing action of overlying gaseous matter on the sun. On the other hand, an accelerated development of such an active region on the sun causes an upheaval of the measured solar heat even when it is situated far from the apparent centre of the luminary, if the activity is not old enough to have sufficient gaseous screens over it. The general level of the solar constant, outside the temporary or short-lived fluctuations due to the above mentioned cases, is sensibly steady and not governed by the apparent circumstances of the solar surface, except in very recent times when the solar activity became weak and solar constant fell possibly as a consequence.

1. Introduction.

In 1913, Dr. C. G. Abbot, Director of the Astrophysical Observatory of the Smithsonian Institution, announced the discovery[2] of the actual variation of the solar heat due to some unknown causes possibly *in* the sun, or at least of trans-terrestrial nature. Subsequent observations of the sun's heat by him and others, made simultaneously at different places, confirmed the discovery. Since some time ago Dr. Abbot has been

engaged in finding connections between the above-mentioned variations of the sun's heat and the apparent phenomena on the solar disc, being led to some suggestions recently. Mr. H. H. Clayton's paper[3] was one of the earliest on this subject. In it he described the idea on which he worked and gave some results showing an increase of the solar constant at the time when a sun-spot comes to some definite points on the apparent solar disc. Last July, when the writer met Dr. Abbot at the Mountain Laboratory of the Mount Wilson Observatory, the latter confided to him his "four principles[4]":

(1) New or increased spots are connected with higher values of the solar constant,

(2) Central spots result in lower values of the solar constants on the next day,

(3) Great activity of hydrogen, Hα, is connected with high values of the solar heat, and

(4) A long quiet period of the solar surface is connected with falling values of the solar constants.

These were the most promising ideas he had obtained in his study up to that time. In a letter dated August 14, 1923, Dr. Abbot wrote to the writer, commenting on (2) of the "four principles", as follows: ".....There are a few cases of radiation minima for which such transits were not observed. It is hoped that some of these will be explained by transits of calcium flocculi disturbances."

The writer started his investigation at the end of July last. Although Dr. Abbot's "four principles" were not apparently consistent with each other, they were of course very precious suggestions to the writer.

2. The Materials Used in the Present Investigation.

(a) *Values of solar constants.* The solar constant was observed more or less continuously at Washington, D. C., during 1902–1907. Since 1905, every summer, except 1907, has been spent at a station on Mount Wilson to observe the constant continuously during each period. In September, 1918, the Calama Station was opened in Chile, South America, with greatly improved equipments, and since then strictly continuous observations of the solar heat have been made there all the year round. In August, 1920, the station was removed to Montezuma, Chile, in the hope of getting better local conditions besides the better instruments, while a northern station of a similar kind was established in October of the same year at Harqua Hala, Arizona, U. S. A. At the present time,

the two stations, Montezuma and Harqua Hala, are rendering splendid services by continuous observations, both enjoying exceptional series of clear weathers every year.

All the observations mentioned above are made by the staff of the Astrophysical Observatory of the Smithsonian Institution, Washington, D. C., and are regularly published in the ANNALS of the same Institution or in the current numbers of the MONTHLY WEATHER REVIEW of the U. S. Department of Agriculture. A series of small corrections is necessary in some recently published values of the solar constant according to Dr. Abbot's special review of the original observations and their reductions. These corrections are not yet published[5], and Dr. Abbot gave them to the writer in MSS.

The writer used all the data of the solar constants which were observed at Montezuma and Harqua Hala, beginning with the earliest days of their services and coming as far as the latest date available. As told by Dr. Abbot himself, these are the best of the serial observations of the solar constants. As a check on the final results of the present investigation, the writer used the data of the observations made at Calama.

The whole data used here are tabulated in Table I below. The first column gives the date, the second the values reduced from the observations at the southern stations, either Calama or Montenzuma, and the third the similar values from Harqua Hala station. The fourth column gives the weighted mean values of the solar constants. The unpublished corrections are applied already. So that these parts of the table are the reproductions of the corresponding data which were already published in the Annals of Astrophysical Observatory of Smithsonian Institution, Vol. IV, pp. 148–158, and the Monthly Weather Review, 1923 February, pp. 77 ff., except the following two points:

i. Unpublished corrections applied,

ii. The very latest values of the solar constant during the early part of 1923, just came to hand from Harqua Hala station.

(b) *Spectroheliograms.* Since the summer of 1915 three kinds of spectroheliograms have usually been obtained every clear day at the Mount Wilson Observatory with its 60-foot Tower Telescope. These are of the calcium K_2 line, the hydrogen C line, and the calcium prominence, which have all been used in the present investigation to give all possible ideas of the solar activity at any date. When some special sun-spots appeared, it was the usual policy of the Mount Wilson observer to obtain large-scale photographs of the region of the solar disc with the hydrogen C line, which have been particularly valuable sources of information of the

Table I.*

Daily Values of the Solar Constants.

Date	Calama Station		Harqua Hala Station		Mean Value	Calcium Area
	Value	Weight	Value	Weight		
1918						
July 27	1·944		——	——	1·944	5·5
28	1·901		——	——	1·901	5·2
29	1·899		——	——	1·899	5·0
30	1·929		——	——	1·929	5·6
31	1·932		——	——	1·932	5·0
August 1	1·945		——	——	1·945	5·2
2	——	——	——	——	——	5·5
3	1·966		——	——	1·966	5·5
4	1·948		——	——	1·948	5·0
5	1·954		——	——	1·954	5·0
6	1·972		——	——	1·972	——
7	1·948		——	——	1·948	3·5
8	1·955		——	——	1·955	3·0
9	——	——	——	——	——	3·0
10	1·954		——	——	1·954	3·0
11	1·965		——	——	1·965	4·0
12	1·959		——	——	1·959	4·5
13	——	——	——	——	——	5·0
14	1·925		——	——	1·925	5·0
15	1·947		——	——	1·947	——
16	1·987		——	——	1·987	5·5
17	1·888		——	——	1·888	5·3
18	——	——	——	——	——	5·3
19	1·948		——	——	1·948	5·2
20	1·940		——	——	1·940	5·5
21	1·995		——	——	1·995	5·5
22	1·953		——	——	1·953	5·6
23	1·979		——	——	1·979	6·0
24	1·932		——	——	1·932	5·0
25	1·968		——	——	1·968	4·5
26	1·949		——	——	1·949	4·5
27	1·955		——	——	1·955	——
28	1,894		——	——	1·894	5·0
29	1·954		——	——	1·954	4·8
30	1·953		——	——	1·953	——
31	2·018		——	——	2·018	5·0
Sept. 1	1·980		——	——	1·980	5·0
2	——	——	——	——	——	4·7

* This is merely a specimen page of the original MS of the present paper. The values of the solar constant have been published:

up to August, 1920.....Annals of the Astrophysical Observatory, Smithsonian Institution, Vol. IV, pp. 149–152;

August, 1920, and after.....Smithsonian Miscellaneous Collections, Vol. 77, No. 3, (February, 1925);

so that I omit here the main part of the MS–Table. The last column of the present Table I, giving the estimated area of Calcium flocculi in the daily spectroheliograms of Mount Wilson Observatory, is my own; but, as I recently found that these values are quite parallel to those of "S. M." series of flocculi area given in Boletin Mensuel del Observatorio del Ebro, I have omitted them too.—*Author.*

sun's phenomena in the present work.

These spectroheliographic observations are of routine character at the Observatory, and the plate number of the running series exceeded 8800 in the end of June 1923, all the plates being preserved carefully in the basement of the Observatory Office, although they have not yet been published in any regular form.

(c) *Sketches of sun-spots and their magnetic polarities.* Since in 1908 Dr. Hale discovered the magnetic field in the sun-spot with the Zeeman Effect, this kind of solar observation has been one of the most characteristic works carried out at the Mount Wilson Observatory. In 1915 Mr. F. Ellerman and others started a routine work of this line, and since 1917 a beautiful series of records of every sun-spot and its magnetic polarity on every available day has been preserved in a uniform manner. Brief summaries of them have been published regularly in the current number of the Publications of the Astronomical Society of the Pacific since 1920. The full details will shortly be published in a special series of the Mount Wilson publications.

The above yet unpublished data were used in the present investigation to trace the phases of local activities of the solar surface. Generally the seeing of the solar image is very important in observing the magnetic field intensities and the fine details of the sun-spot. When the seeing is 2 or under in Ellerman's scale, the observed field-intensity reveals merely its *lower limit.*

3. Nature and Phases of the Heliographic Activity.

It has become necessary in the present investigation to distinguish the "phase" of the solar activity. Generally speaking, an activity of the sun is represented by the existence of some apparent local disturbance of the uniformity of the solar surface besides the "darkening" of radiation toward the limb. Sun-spots are the most striking disturbances on the sun, and have long been known. The early days of the spectroheliograph revealed that the sun-spot is always surrounded by calcium flocculi, and moreover it is very often the case that many calcium flocculi are found quite independent of the appearance of any sun-spot. A patch of the flocculi usually survives the sun-spot which it surrounded. Also a new spot is born generally from pre-existent flocculi, so that the calcium flocculi are to be considered as the true home of a sun-spot, and are the most effective indication of the local solar disturbance on the disc, while the spot is only the "peak" of the local activity shown more or less strikingly.

Spectroheliograms show the dark filaments of absorbing hydrogen on the sun when observations are made with the H*a*-line. Generally the structure of a hydrogen filament is streaky and curved, and the total length sometimes reaches the diameter of the solar disc. The orientation of the filament is not limited like that of the calcium flocculi, but occurs at any point of the sun. A few years ago, Mr. F. Ellerman identified the filament with the solar prominence, and the prominence was proved some time ago by Dr. Hale to be helical in its usual structure. In 1917 O. J. Lee[5] published a paper which disproved a definite relation between the sun-spot and the prominence. But, the writer's experience connected with the present investigation shows some very significant relations existing between the sun-spot and the hydrogen filament, not to speak of the "prominence." Generally speaking, a filament is connected to some group of spots or flocculi, although it is often apparently distant from them.

The typical development of a local solar activity, according to the writer's observation, might be sketched as follows: At first there is a small patch of calcium flocculi on a certain point of the solar disc. These flocculi come out of a minute pore of the sun's granulation more or less suddenly, and the patch becomes conspicuous both by the extension of its area and also by the increase in its brightness. At a certain period of its development, a point more or less central to the flocculi area becomes darker, and there comes a sun-spot, very often followed by another spot, within the area of the same flocculi. Usually the preceding spot maintains its circular form and continues its development, becoming larger up to a certain limit, the field strength of the magnetic polarity being intensified proportionately or otherwise. The following spot is generally unsteady in its structure, and apt to break up into several components, distributed over a wider area. Rarely some of the components retain the original intensity of the magnetic field for an interval, but usually the magnetism is weaker in any components of this following spot. The opposite character of the polarity between the preceding and the following spots within the same flocculi was discovered by Dr. Hale some years ago. As the spots develop further, the field intensity of the magnetic polarity reaches a maximum, and then begins to fall, while at the same time the whole system of the activity "degenerates", extending still further the total area of the all-surrounding flocculi with many indentations in its outlines. Absorbing hydrogen filaments usually develop right over the area of activity when it is reaching the climax of its development. From the beginning the filaments are curved and sometimes twisted, more or less concentrically surrounding the sun-spots at least partially. These filaments

tend to recede afterwards from the central portions of the activity probably because of some expelling forces, and sometimes reach points as far as a considerable fraction of the apparent solar diameter. Very often a well-extended filament is seen stretching 20′ or more in arc over the solar surface, and looks like quite an independent feature from others on the hydrogen spectroheliogram. But the finer structures of the details of a filament are streaky and their partial curvatures are always concave to the same side of the body, on which side we can find some areas of great former disturbances in the form of sun-spots or calcium flocculi. Such is an outline of a typical development of a sun-spot and the appearances related to it. Actually there are many varieties of appearances as we see on the solar disc visually or photographically. But all are special cases of the typical one just sketched. In an ideal case we can distinguish the *phase* of the development of the solar activity by inspecting the spectroheliograms of calcium and hydrogen lights, namely in the following three ways :

(i) Sun-spot structure in its details. If compact, then it is young, and if decomposed into several members then it is so much degenerated.

(ii) Calcium flocculi. If the occupied area is extended, and if the outline is zigzag-shaped, it is so far developed.

(iii) Hydrogen filament. If it is seen very near the area of activity, more or less crooked or just coming out radially from the spot centre, then the whole system is of young development. If the filament is more or less detatched from the activity region surrounding it from a distance, then the development is already well advanced. If some long series of filaments are stretching over the solar surface and not directly connected with any particular spots, this possibly represents a story of some activity of former days.

There is one more way of distinguishing the phase of the sun-spot activity, that is by

(iv) Serial Observations of the magnetic field-intensity of the region as carried at by Mount Wilson observers.

I admit that the idea of the *phase* (or *age*) of solar activity is too new for a universally applicable quantitative definition of that activity to be possible by its means ; it yields many ways of definition from diverse points of view. The writer assumes here, as a preliminary idea, that the degree of the activity can be represented by the intensity of the magnetic field of the sun-spot given by the Zeeman Effect. It is interesting to the

writer that Dr. S. B. Nicholson[7] of Mount Wilson has obtained a significant relation between the sun-spot and its magnetic field intensity.

4. Decipherment of Variation of the Solar Constant in Terms of the Heliographic Activity.

In the early days of the writer's present investigation, he attempted to obtain some relation between the general tendencies of the solar constant and the calcium flocculi. For this purpose, eye-estimations of *apparent effective area** of calcium flocculi on daily spectroheliograms were made, which are given in the last column of Table I and plotted on cross-section papers so that they might be compared with the similar curves of the solar constant, both being in the same scale with regard to the abscissae representing time. The direct comparison was not promising at first, only sometimes showing parallelism positively as well as oppositely. During this stage of examination of the materials, the writer sometimes found very peculiar habits of quite a temporary character in the curve of the solar constant. Usually when a well developed area of solar activity comes to the apparent centre of the solar disc, a sudden drop in the solar heat occurs; the effect being in some cases for one or two days, and in others much longer. This is just what Dr. C. G. Abbot suggested in the second of his "four principles". But in some cases such near approach of the spot area to the solar centre does not bring the corresponding depression of heat: sometimes the curve of the solar constant remains calm, showing no effects at all, while sometimes the heat curve turns *upward*! Then the writer took some typical examples of the three cases: the falling, the rising and the unalteration of the solar heat corresponding to a central passage of a sun-spot. A close examination of all the available materials and exhaustive comparison of the three cases gave the following general conclusions:

(A) If the sun-spot is in its developing stage of activity into a higher order at the time of its central passage across the sun's apparent disc, the solar heat goes up.

(B) If the sun-spot is already well developed, and especially when it is in the degenerating stage of activity, then the solar heat falls down.

(C) The sun-spot which does not produce any apparent effect on the solar heat by its central passage across the sun is one which is

* *Apparent Effective Areas* of Activity are estimated here by putting relatively much weight on those near to the centre of the solar disc and less weight on those at the limb.

in its full development of activity, just not too young and yet not too old, so that it is near the climax of activity.

With these three principles, the writer was able to decipher many temporary variations of the solar constant. The following are some examples illustrating typical cases. Some of the most notable examples showing the *increasing* activity accompanying heat evolution are given in the following tables. See, also, the photographs in Fig. 1 illustrating these examples.

EXAMPLE I.

Table II.

Activity of Sun-spot No. 1840 (Mt. Wilson)

Date	Magnetic polarity		Solar Constant	K_2–Flocculi estimated.
	Intensity	Remarks		
1921 April 19	V ? R ?	at limb	1·920	3·5
20	V 7R 3		1·968	3·3
21	?	no obs.	1·921	3·0
22	V 4R 7		1·956	3·0
23	——	extinguished	1·919	3·0
24	——	invisible	1·933	3·0
25	V 5	revived!	1·946	——
26	V 4R 5		1·944	2·5

Solar meridian passage: April 24, 1921.

EXAMPLE II.

Table III.

Activity of Sun-spot No. 1873 (Mt. Wilson)

Date	Magnetic polarity		Solar Constant	K_2–Flocculi estimated.
	Intensity	Remarks		
1921 July 8	V12R 4		1·937	3·2
9	V18R 6		1·959	3·3
10	V26R10		1·964	3·2
11	V17R12		1·936	2·7
12	V17R 8		——	2·0
13	V17R14		1·951	2·0
14	——	weak	1·954	2·2

Solar meridian passage: July 8, 1921.

EXAMPLE III.

Table IV.

Activity of Sun-spot No. 1915 (Mt. Wilson)

Dete	Magnetic Polarity		Solar Constant	K_2–Flocculi estimated
	Intensity	Remarks		
1921 October 24	R24V23		——	3·0
25	R24V22		1·942	3·0
26	R25V22		1·952	3·0
27	R23V18		1·946	2·7
28	R27V25		1·962	2·5
29	R33V20		1·955	2·4
30	R28V18		1·962	2·0
31	R27V10		1·957	2·0
November 1	R26		1·949	1·5
2	?	limb	1·955	1·0

Solar meridian passage: October 27, 1921.

In these tables showing EXAMPLEs, the magnetic intensity, for examples, V7R3, means that the *maximum* intensity of the magnetic field of the preceding spot is 7 (ca. 700 gausses in Ellerman's scale) in the counter-clockwise vortex and the *maximum* intensity of the field of the following spot is 3 (ca. 300 gausses in Ellerman's scale) in the clockwise vortex; the columns of the Solar Constant and the K_2–Flocculi are simply reproduced from the Table I of the present paper for direct comparison. The case of Sun-spot No. 1873 is conspicuous for the rapid rising of the solar constant, which attained its temporary peak on July 10 as seen in Fig. 1. In the case of No. 1840, the spot made its appearance first on April 19 when it was seen near the east limb and numbered as No. 1839; this spot was extinguished on the solar disc by April 23 and suddenly *revived* later as No. 1840 on the first day after its meridian passage—a rather unique case—and the outburst of the new energy is shown in the rising of the solar constant at that time. No. 1915 made the solar heat go steadily upward, and then disappeared behind the visible hemisphere of the sun by its rotational effect; while after a due interval the spot came back into sight from the east limb in the middle of November, and passed the meridian on Nov. 23, by which time it had reached its full deveolpment and evolved conspicuous hydrogen filaments and consequently the cutting out of the solar heat thereby was very conspicuous in the observations of the solar constant.

Example of *temporary drops* in the solar constant simultaneous with the meridian passage of some solar activity area are numerous. The following cases are given here:

EXAMPLE IV.

Table V.

Activity of Sun-spot No. 1809 (Mt. Wilson)

Date	Magnetic Polarity		Solar Constant	K_2–Flocculi estimated
	Intensity	Remarks		
1921 February 13	V 5		1·960	2·5
14	V 5		——	——
15	V 5		1·960	2·0
16	V 5		1·961	2·2
17	——		1·955	——
18	V26R28		1·948	2·5
19	V23R19		1·926	3·0
20	V24R24		1·932	3·4
21	V20		1·959	——
22	V20		1·948	3·3
23	V18R16		1·943	3·0
24	?	limb		

Solar meridian passage: February 18, 1921.

EXAMPLE V.

Table VI.

Activity of Sun-spot No. 1842 (Mt. Wilson)

Date	Magnetic Polarity		Solar Constant	K_2–Flocculi estimated
	Intensity	Remarks		
1921 May 8	?		1·955	2·8
9	V17R14V12		1·944	3·0
10	R20V25		1·962	3·2
11	R24V33		——	3·5
12	R28V30		1·948	3·8
13	R34V32		1·964	4·0
14	R54V30		1·941	4·0
15	R31V23		1·934	——
16	R28V26		1·938	——
17	——	Cloud.	——	——
18	?		1·957	——
19	?		1·963	——

Solar meridian passage: May 14, 1921.

EXAMPLE VI.

Table VII.

Activtiy of Sun-spot No. 2014. (Mt. Wilson)

Date	Magnetic Polarity		Solar Constant	K_2-Flocculi estimated
	Intensity	Remarks		
1923 April 19	V ? R ?		——	2 0
20	V24R19		1·940	1·5
21	——	cloud	1·942	——
22	——	cloud	1·917	1·4 ? ?
23	V11R 5		1·908	1·5
24	V 9R 3		1·920	1·3
25	V12R 5		1·932	1·0
26	V12R 5		1·948	1·3
27	V10R15		1·924	1·2
28	?		1·927	1·3

Solar meridian passage: April 23, 1923.

Spot No. 1842 returned after a rotation of the luminary to the visible hemisphere as No. 1852, which passed the central meridian and effected heat absorption again. No. 2014 had disappeared by the time when its next return was due, but the calcium flocculi survived it and apparently effected a reduction in the solar heat at the time of its meridian passage on or about May 20.

The cases in which the *plus* and *minus* effects on the solar constant were almost equal and consequently balanced are relatively rare. The following are good examples:

EXAMPLE VII.

Table VIII.

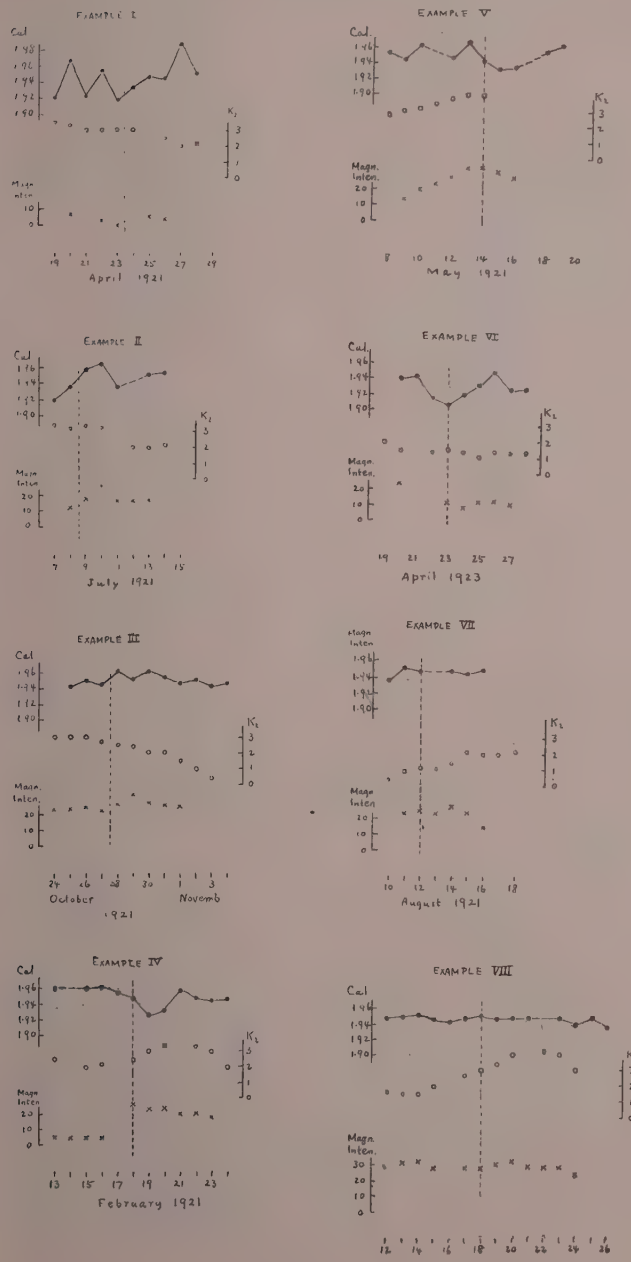
Activity of Sun-spot No. 1889 (Mt. Wilson)

Date	Magnetic Polarity		Solar Constant	K_2-Flocculi estimated
	Intensity	Remarks		
1921 August 11	R24V11		1·951	1·0
12	R25V 7		1·946	1·2
13	R23V 3		——	1·1
14	R28		1·946	1·5
15	R24		1·943	2·2
16	R14		1·947	2·0
17	?		——	2·0
18	?		——	2·2

Solar meridian passage: August 12. 1921.

Fig. 1.

Remarks: Dotted vertical lines show the dates of central meridian passages of the sun-spots.



EXAMPLE VIII.

Table IX.

Activity of Sun-spot No. 1903 (Mt. Wilson)

Date	Magnetic Polarity		Solar Constant	K_2–Flocculi estimated
	Intensity	Remarks		
1921 September 12	?		1·947	1·6
13	R31		1·949	1·5
14	R32		1·952	1·5
15	R28		1·944	2·0
16	R ? V 2		1·941	——
17	R28		1·946	2·6
18	R28V ?		1·950	3·0
19	R30		1·943	3·3
20	R32V ?		1·945	4·0
21	R29V 3		1·946	——
22	R28		——	4·2
23	R28		1·946	4·0
24	R24		1·939	3·0
25	?		1·946	——

Solar meridian passage: September 18, 1921.

No. 1903 returned to this side of the sun in the next month, and passed the solar meridian on October 16 effecting a conspicuous drop in the solar constant.

Some of the best plates of the Mount Wilson spectrograms, taken with the H*α*–light, and sometimes with direct light, showing the characteristic features in connection with the present problems, are given in the appended Plates.

The physical meanings of these principles are probably as follows: When the solar activity is increasing, it means that some accelerating forces are acting upon the region, and consequently radiating energy is evolving, which affects the total heat received by our terrestrial instrument. But, along with further development of the activity area, hydrogen and other gaseous matter are evolving out of the region by mechanical or some other form of expulsion, and when these gases become predominant *over* the area absorption of out-going energy takes place, which absorption continues to prevail, sometimes becoming more powerful, even after the original active source (spots) has begun to die away. Hence, the effect under principle (1) is due to some direct action of the active area upon

the earth, and that under (2) is due to absorption by overlying gases. When these two agencies are working at the same time, the result upon our terrestrial pyrheliometer is determined by the relative efficiency of them. The phenomena under principle (3) are the result of the two agencies, just balancing so to speak, which is the reason why it occured when the activity was in full development.

Later, upon examining many points of interest of the curve of the solar constant, keeping in mind the three principles as above, the writer found that it was not always necessary for an area of activity to come just upon the centre of the solar disc to affect a rise in the solar constant if its activity is increasing. It is really often the case that such kind of activity brings about the same result when it is situated rather distant from the apprent centre of the sun. Generally speaking, an increasing activity taking place upon the *visible* part of the solar surface is enough to produce a rise in the solar constant if it is sufficiently strong to do so. A new birth of a sun-spot is of course a special case of this type. When a sufficiently strong activity develops just at the time when it comes within the visible limb of the solar disc it may have the same effect: this type will explain the "limb effect" detected some years ago by Mr. H. H. Clayton.* Statistically, however, the number of cases where the absorption take place as shown in the solar constant curve is far greater than that of those where a rising in the solar heat is observed. This should be rightly justified from the physical meaning of the phenomena, because the rising stage of an area of activity upon the sun is much shorter in life than that of its falling stage. The cases, in which the two agencies opposite to each other just balance, are the least in number, because these must be really exceptional cases.

In No. (3) of his "four principles", Dr. Abbot mentions some connections between great activity of hydrogen and high values of the solar heat. The meaning of "hydrogen activity" is a little ambiguous. If he means by that phrase some energetic evolution of hydrogen filaments newly developing from the activity centre of a sun-spot, the writer's experience with the spectroheliograms in the present investigation confirm the statement eloquently. If he means by the term only splendid apparitions of big filaments over the visible surface of the sun without any rapidity of changes of structure, his principle cannot conform with the present study, because in such cases the writer often sees more or less absorption predominant upon the sun by the overlying gases.

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* loc. cit.

Sometimes the calcium flocculi without any spots in them affected the solar constant, which is a direct proof of the comment on No. (2) of his "four principles" by Dr. C. G, Abbot. Downfalls of solar heat on

July 26–27, 1921,
July 5–7, 1922,
August 15–10, 1922,

are beautiful examples of the effects of the flocculi when they passed the meridian on those occasions. Rise in the solar constant was also often observed as due to effects of new births of flocculi, those on the dates:

April 16–18, 1922,
October 7–9, 1922,
August 24–26, 1922,

being examples of this.

Generally the date of the minimum of the solar constant due to the meridian passage of the local activity on the sun is one day later than the date of the passage. But this is not strictly the case for all of them. Sometimes the minimum comes just on the date of the meridian passage, and sometimes it comes a short while *after* that time apparently. Accurate determination of this kind of phase difference cannot be made from the present materials, because the time of observing the solar constant is not suffiiciently given for accuracy.

The writer has met several cases in which the solar surface was quite free from any activity upon it. In such cases the fluctuation of the solar constant is generally inappeciable, but it cannot be easily concluded whether the solar constant is usually falling down or going up during such a period. There are varieties among them. For example, in the following intervals the solar surface was quite calm:

May 13–16, 1922,
October 22–28, „ ,
November 22–27, „ ,
December 14–21, „ ,
January 5–27, 1923,
February 17–28, „ .

In four of these cases in 1922 the solar heat was going *up*, sometimes with rapidity. In examining the calcium spectroheliograms carefully, the writer detected in these cases some signs of flocculi just evolving from the photospheric background. On the other hand, the two cases in 1923 showed a falling down of the solar heat, and similar examinations of the Plates revealed some traces of old activity on the sun's surface.

Statistically speaking, however, a quiet period of the solar surface is connected with steadiness of the solar radiation, and if this means the absence of new sources of energy on the surface, with possible remnants of gaseous clouds of former days, it may result in a decline of the total heat anyhow.

5. Study of the Run of Mean Values of the Solar Constant.

The general run of curves, that is, of the solar constant and of the effective flocculi area plotted in the same form upon cross-section paper, short-lived variations being disregarded, was examined with a view to seeing whether there was any parallelism between them. No generalization in the question proved to be possible: sometimes they ran quite parallel, the rising and falling of the one corresponding to similar characters of the other, while sometimes the run was opposite in mutual correspondence, and in other cases there was no relation at all. From the curves extending over an interval of six years, the writer obtained the result shown in the following Table:

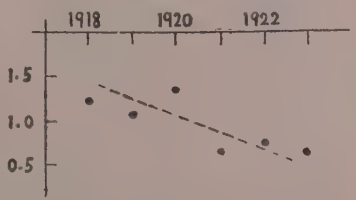
Table X.

Number of Days of Relationship.

Year	1918	1919	1920	1921	1922	1923
Parallel Run	44	116	168	99	107	28
Opposite Run	35	111	121	162	136	42
Ratio	1·24	1·08	1·37	0·61	0·75	0·66

It is to be remembered that the number of days of observations here used in the years 1918 and 1923 is not complete, being 155 and 142 days respectively. In the third row the ratio of the two kinds of runs, in the sense of *Parallel Run to Opposite Run*, is given. Stress need not here be laid upon the numerals strictly, but the acompanying Figure 2 shows the general tendency of the Ratio. As a rough suggestion, it may be stated that the sense of parallelism is changing, or at least is not the same, from the early part to the latter part of the interval. The last maximum of sun-spot activity was in 1917, as was widely observed,

Fig. 2.



so that the years 1918 and 1919 are still years of strong activity while in the later years the sun is becoming more and mare calm. Bearing in mind these circumstances existing *in* the sun, the writer is led to reflect that in the earlier years of the present interval the solar effect upon the solar constant was predominating in its energy-radiating power, while in the later years the effect of gaseous absorption was controlling the general field, so to speak.

Strictly continuous observations of the solar constant have been made since September 1918, but at the Mount Wilson Station of the Astrophysical Observatory of the Smithsonian Institution, observations of the same constant have been continued almost uninterruptedly every summer since 1905; so that we have here a precious series of materials for the study of the annual run of the solar constant here if that study be restricted to summer. The following Table XI gives all available values of the mean solar constant of every *August* since 1904 observed at several stations solely or simultaneously.

Table XI.

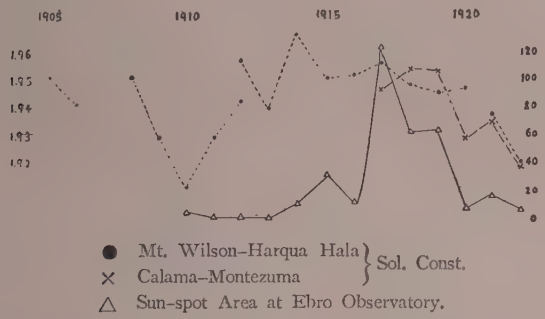
Solar Constant and Sun-spot Area Every August.

Year	Solar Constant			Sun-spot Aaea
	Mt. Wilson	Calama-Montezuma	Harqua Hala	
1905	1·951	——	——	——
1906	1·941	——	——	——
1907	——	——	——	——
1908	1·951	——	——	——
1909	1·930	——	——	——
1910	1·912	——	——	7·1
1911	1·930	——	——	0·8
1912	1·957	——	——	0·2
1913	1·940	——	——	0·3
1914	1·944	——	——	11·0
1915	1·951	——	——	31·2
1916	1·952	——	——	12·5
1917	1·956	1·947*	——	123·8
1918	1·948	1·954	——	61·8
1919	1·945	1·953	——	63·5
1920	1·947	1·929	——	7·2
1921	——	1·935	1·937	19·3
1922	——	1·918	1·920	4·8

* In 1917 a series of observations of the solar constant was made at Hump Mountain, N. C., with the instrument which was transported to Calama in the following year.

In Figure 3 the values of Table XI are plotted, together with the sun-spot observations (in terms of heliographic area per square millimeter of the solar disc) made at the Ebro Observatory and published in its

Fig. 3.



BOLETINs. We see here that in the early years there is no apparent relation between the heat and spots of the sun, but in the later years there is a decided parallelism between them. If we reduce the apparent relations of the two phenomena to numerical form from the best and unform materials obtained at the Calama-Montezuma and Ebro observatories, we obtain :

$$K = 1{\cdot}923 + 0{\cdot}000469\,x,$$

where K is the solar constant and x the Ebro Sun-spot area.

Next, the writer computed monthly the mean values of the solar constant from the data obtained at the Calama–Montezuma and the Harqua Hala station, and at the same time he obtained similar values of the estimations of the effective area of the calcium flocculi made by himself from the Mount Wilson spectroheliograms, all these values being given in Table XII below :

According to Dr. C. G. Abbot, the best of the determinations of the solar constant made under his supervision are those made in the interval from August 1920 up to the latest date of 1923 at the Montezuma and the Harqua Hala stations. The writer rounded the *weighted* mean values of them, still further to get a smoother curve, by taking the means of every consecutive three values, and these are given in the fourth row of Table XII. All the numerical values of Table XII are a'so plotted in Figure 4 given here.

An annual periodicity of the solar constant, which was now computed from the data up to the end of 1921, is apparent, because the rapid fall

Table XII.

Monthly Mean Values of the Solar Constant together with those of Calcium Area.

	Epoch	Jan.	Feb.	Mar.	April	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.
1918	Calama	—	—	—	—	—	—	—	1·954 27	1·944 18	1·935 24	1·942 23	1·959 19
	Harqua Hala	—	—	—	—	—	—	—	—	—	—	—	—
	Calcium Area	—	—	—	—	—	—	—	4·8	4·5	5·4	5·0	5·3
1919	Calama	1·944 19	1·948 20	1·941 16	1·952 27	1·940 27	1·955 22	1·954 27	1·953 30	1·939 28	1·952 20	1·953 25	1·952 24
	Harqua Hala	—	—	—	—	—	—	—	—	—	—	—	—
	Calcium Area	4·8	5·2	5·0	4·5	4·8	5·2	5·1	5·1	4·9	4·1	4·0	4·6
1920	Calama–Montezuma	1·964 25	1·956 19	1·946 29	1·952 30	1·953 29	1·939 23	1·945 21	1·929 27	1·947 25	1·945 21	1·948 26	1·956 22
	Harqua Hala	—	—	—	—	—	—	—	—	—	1·944 14	1·953 11	1·937 5
	Weighted Mean	—	—	—	—	—	—	—	1·929	1·947	1·945	1·950	1·952
	Rounded									1·940	1·947	1·949	1·955
	Calcium Area	3·8	4·4	4·0	2·9	3·3	3·5	2·8	1·9	2·0	3·7	3·7	3·6
1921	Montezuma	1·955 9	1·956 7	1·946 13	1·944 16	1·943 13	1·939 17	1·949 18	1·935 1	1·953 5	1·951 12	1·949 15	1·952 12
	Harqua Hala	1·968 12	1·947 19	1·941 14	1·944 17	1·951 17	1·935 21	1·934 12	1·937 13	1·543 26	1·944 22	1·958 19	1·949 11
	Weighted Mean	1·962	1·950	1·943	1·944	1·948	1·937	1·943	1·937	1·944	1·947	1·954	1·950
	Rounded	1·958	1·952	1·946	1·945	1·943	1·943	1·939	1·941	1·943	1·948	1·950	1·949
	Calcium Area	3·0	2·6	2·9	2·8	2·5	2·5	2·8	2·1	2·1	2·2	2·0	1·8
1922	Montezuma	1·947 19	1·947 10	1·938 14	1·926 12	1·917 6	1·913 11	1·910 8	1·918 10	1·934 16	1·932 10	1·929 8	1·909 2
	Harqua Hala	1·940 15	1·946 12	1·926 15	1·921 20	1·929 22	1·920 24	1·911 20	1·920 11	1·909 22	1·919 29	1·910 24	1·921 16
	Weighted Mean	1·944	1·947	1·931	1·923	1·926	1·918	1·911	1·919	1·914	1·922	1·915	1·920
	Rounded	1·947	1·941	1·934	1·927	1·923	1·918	1·916	1·915	1·918	1·917	1·917	—
	Calcium Area	2·2	2·2	2·7	2·1	1·8	1·4	1·7	1·1	1·0	1·6	1·2	1·3
1923	Montezuma	—	—	—	—	—	—	—	—	—	—	—	—
	Harqua Hala	1·911 17	1·903 18	1·908 22	1·923 19	1·919 21	—	—	—	—	—	—	—
	Calcium Area	1·1	1·1	1·0	1·4	0·7	1·2	—	—	—	—	—	—

of the general curve in 1922 seems too unusual to be treated without consideration of some special kind. The annual term in the sense of deviations from the mean constant during the three and a half years, 1918–1921, is as follows:

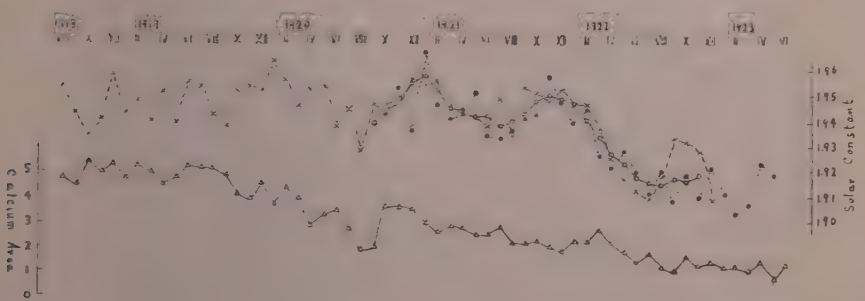
Table XIII.

Annual Variation of the Solar Constant.

Month	Mean Solar Constant	Rounded from consecutive three values
January	+·009	——
February	+·003	+·002
March	−·005	+·000
April	−·0021	−·002
May	−·001	−·001
June	−·004	−·002
July	−·001	−·003
August	−·005	−·004
September	−·005	−·004
October	−·003	−·002
November	+·002	+·001
December	+·005	+·005
(January)	(+·009)	+·006

The January value in the last row is repeated for convenience. Here the true cause of this annual variation of the solar constant is not easily identified; it may be due either to the effects of the sun's declination or to that of the sun's distance, or it may come from some instrumental origin as well as from the methods of reduction of the original observation.

Fig. 4.



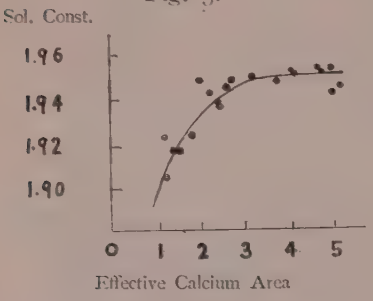
Now the above-obtained values of the annual term of the solar constant were applied as *corrections* to individual monthly means to bring them to normal values, which were then reduced to another series of mean values for each quarter year, the process being extended to the latest values of 1923. The result is shown in Table XIV below:

Table XIV.

Mean Normal Solar Constant and Calcium Area in each quarter year.

Month	1918		1919		1920		1921		1922		1923	
	Sol. Const.	Calcium	Sol. Const.	Calcium	Sol. Const.	Calcium	Sol. Const.	Calcium	Sol. Const.	Calcium	Sol. Const.	Calcium
January–March	—	—	1·942	5·0	1·953	4·1	1·949	2·8	1·938	2·4	1·905	1·1
April–June	—	—	1·951	4·8	1·950	3·2	1·945	2·6	1·924	1·8	1·923	1·1
July–September	1·953	4·6	1·952	5·0	1·942	2·2	1·939	2·3	1·918	1·3	—	—
October–December	1·944	5·2	1·951	4·2	1·948	3·7	1·949	2·0	1·918	1·4	—	—

Fig. 5.



These values are diagrammatically shown in Figure 5. It is apparent that calcium flocculi of a value of more than 3 in the present scale keeps the solar heat sensibly constant, while the effect is marked when it is below 3 in the scale.

6. Summary and Conclusion.

For the purpose of finding the influence, if any, of heliographic activity upon the solar constant, a considerable amount of data was examined, those data comprising the spectroheliograms of different kinds and magnetic observations of visible and invisible sun-spots made at the Mount Wilson Observatory, to compare them with the run of the solar constant values obtained at the Astrophysical Observatory of the Smithsonian Institution. The nature of the phenomena is pretty complex. Temporary or short-lived effects of the solar activity upon the solar constant are divided in two kinds according to the life-history of the area of activity concerned. An area of increasing activity on the sun makes

the solar heat rise, the apparent heliographic position of the area not being of essential importance. An activity area in decay, on the other hand, makes the observed solar heat drop when it comes sufficiently near to the centre of the apparent solar disc, probably by some screening action of overlying gases evolved from the region. Sometimes the two agencies occur at the same time, naturally in the opposite sence in their effects upon the solar heat, so that a good balance occasionally results between them. For the general relation between the solar activity and the solar heat controlling circumstances of long duration, the case is not always the same for the earlier and the later half of one complete 11-year cycle of solar variation. An apparent conspicuous relation between them is revealed when the sun is generally calm in the cycle.

The present investigation was made by suggestions given to the writer early by Dr. C. G. Abbot, whose keen interests in and careful watch upon the progress of the work encouraged the writer, whose hearty thanks are naturally due to him. It was especially fortunate for the writer that Dr. W. S. Adams, Director of the Mount Wilson Observatory, allowed him to use freely the rich stock of materials accumulated in the course of many years by the staff of the observatory, to whom many thanks are also due.

Issei Yamamoto.

Mount Wilson Observatory,
Pasadena, California, U. S. A.

October 23, 1923.

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1 An abridged report of this investigation was published in Monthly Notices, R. A. S., Vol. LXXXV, p. 71, (1924).

2 Annals of the Astrophysical Observatory of the Smithsonian Institution, Vol. III. p 18.

3 Nature, Vol. **105**, 630, (1921) and Vol. **107**, 108. (1921).

4 Abbot has subsequently published these principles in Proc. Nat. Acad. Sc. U. S. A., Vol. **9**, p. 355, (1923).

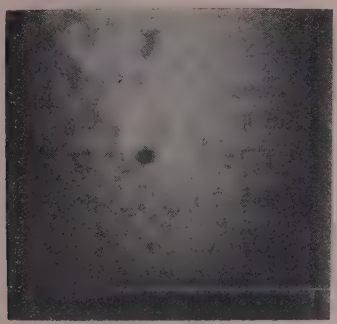
5 The final values with these corrections have recently been published in Smithsonian Miscellaneous Collections, Vol. **77**, No. 3, (1925).

6 Astrophysical Journal, Vol. **45**, p. 206, (1917).

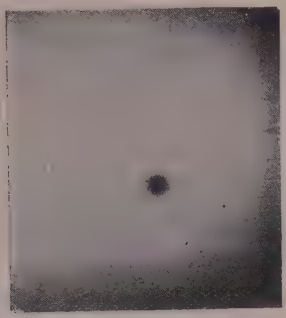
7 Publ. A. S. Pacific, Vol **31**, p. 182. (1919).

Plate I.

Mt. Wilson Sun-spot No. 1823, on March 29, 1921, in *growing* stage.



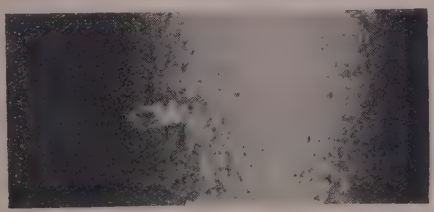
Hα image.



Direct image.

Plate II.

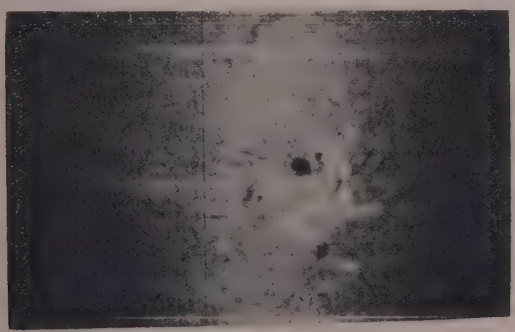
Mt. Wilson Sun-spot No. 1902, on Sept. 11, 1921, just *evolving*.



Hα image.

Plate III.

Mt. Wilson Sun-spot No. 2005, on Dec. 27, 1921, *growing* up.



Hα image.

APPENDIX I.

Recomputation of the Relation between the Solar Constants and the Calcium Flocculi.

On the suggestion of Dr. W. S. Adams, I made two series of computations: first, for partial intervals of the solar constant value, and next, for those of the flocculi area. These results are shown in the accompanying Fig. 6, where the "small circles" represent the values obtained for each interval of flocculi area and the "crosses" those for intervals of solar constant. There, certainly, some kinds of regression are apparent. But, frankly, I was not quite convinced of the matter, partly, from the present computational experiences, and partly from some recollections of similar cases connected with the computation of Table XII of my principal paper dated October last. For, Table XIV and Fig. 5 in the preceding paper show, that the mean solar constant values connected with higher values of the flocculi are, without exception (!), belonging to earlier; while those values connected with the lower flocculi values generally belong to later dates; so that the regression curve in Fig. 6 is not only determined by the solar constant and the flocculi but also by the period.

For a clearer demonstration of the time-effect of the matter, I recomputed the relations which were already shown as cross-series in Fig. 6, and in this case the whole material was first divided into two series, one from the data of 1918–1920 and the other from those of 1921–1923. These two of computations were made separately, and the results are plottod. again, in the same figure (Fig. 6) here. The parallel character of each of the curves is quite clear, suggesting that the regression-form represented by the previous "crosses" is not so definite as might be supposed from its appearance.

Next, I proceeded to repeat similar computations further: but in this case I made them *for each year*. The results are shown in the following Table XV, and are given diagrammatically in Fig. 7.

In each of the annual rows of Table XV, the first row is the mean solar constant, the second, the mean flocculi area, and the last the weight; those values which have too small weights were conveniently combined with neighboring values to furnish data to be shown in Fig. 7. The values for 1918 and 1919 almost coincide; besides, the curves for 1922 and 1923 are as near to each other as those for 1920 and 1921;

Table XV.

Year	<1·896	1·900 −1·909	1·910 −1·919	1·920 −1·929	1·930 −1·939	1·940 −1·949	1·950 −1·959	1·960 −1·969	>1·970
1918	1·885 4·97 7	1·904 5·47 3	1·914 5·50 2	1·925 5·14 10	1·934 4·85 10	1·945 4·92 15	1·953 4·86 19	1·964 5·04 7	1·982 5·29 10
1919	1·898 2·20 1	— — —	1·918 4·83 3	1·926 5·00 13	1·935 5·05 17	1·945 4·85 24	1·955 4·77 41	1·963 4·82 37	1·977 5·04 14
1920	1·880 4·53 3	1·906 1·80 2	1·916 2·70 6	1·924 2·70 19	1·935 3·08 30	1·945 3·07 51	1·953 3·34 50	1·964 3·28 31	1·984 3·65 11
1921	1·871 3·55 2	1·903 3·15 2	1·915 2·63 6	1·924 2·89 13	1·935 2·37 32	1·944 2·48 65	1·954 2·22 41	1·963 2·40 30	1·982 2·43 9
1922	1·880 1·57 15	1·905 1·40 21	1·915 1·67 40	1·922 1·51 46	1·934 1·81 40	1·944 1·94 24	1·954 2·20 6	1·964 0·35 2	— — —
1923	1·884 1·09 8	1·905 0·88 18	1·914 1·08 14	1·925 0·82 17	1·932 0·94 5	1·943 0·83 4	— — —	— — —	— — —

so that these points of curves were finally combined, except those for 1920 and 1921, and reproduced in Fig. 8. (This may not be necessary, because the six curves in Fig. 7 are quite distinct; but anyway, those for 1918 and 1919 are rather convenient when they are united, their differences being insignificant apparently.) The values obtained by these numerical combinations are:

Table XVI.

Years	<1·899	1·900 −1·909	1·910 −1·919	1·920 −1·929	1·930 −1·939	1·940 −1·949	1·950 −1·959	1·960 −1·969	>1·970
1918–19	1·887 4·62 8	1·904 5·47 3	1·916 4·90 5	1·926 5·06 23	1·935 4·97 27	1·945 4·88 39	1·954 4·80 61	1·963 4·86 44	1·979 5·14 24
1922–23	1·881 1·40 23	1·905 1·16 3	1·915 1·52 54	1·923 1·32 63	1·934 1·71 45	1·944 1·78 28	1·955 2·20 6	1·964 0·35 2	— — —

It will be noticed that in both Figs. 7 and 8, the curve are all practically vertical, and only the annual run from large flocculi area to smaller is definitely shown.

Next, I re-computed the values as shown in the "small circle" series in Fig. 6, similarly, for each year. The results are:

Table XVII.

Year	<1·0	1·0 −1·4	1·5 −1·9	2·0 −2·4	2·5 −2·9	3·0 −3·4	3·5 −3·9	4·0 −4·4	4·5 −4·9	5·0 −5·4	>5·5
1918	—	—	—	—	—	1·953	1·945	1·924	1·949	1·939	1·945
	—	—	—	—	—	3·10	3·50	4·03	4·63	5·07	5·66
	—	—	—	—	—	3	2	7	12	30	29
1919	—	—	—	1·927	1·951	1·962	1·968	1·953	1·952	1·950	1·954
	—	—	—	2·10	2·60	3·00	3·50	4·04	4·60	5·08	5·74
	—	—	—	2	2	4	1	16	27	72	26
1920	—	1·936	1·944	1·947	1·946	1·949	1·952	1·948	1·950	1·939	1·952
	—	1·13	1·61	2·16	2·63	3·13	3·55	4·12	4·60	5·09	5·60
	—	18	17	26	24	31	21	21	24	15	6
1921	1·936	1·951	1·951	1·951	1·941	1·943	1·944	1·951	1·891	—	—
	0·53	1·08	1·58	2·11	2·61	3·10	3·59	4·03	4·50	—	—
	6	22	19	46	30	49	18	9	1	—	—
1922	1·922	1·920	1·915	1·925	1·932	1·922	1·941	1·932	1·933	—	—
	0·55	1·13	1·59	2·13	2·55	3·10	3·50	4·10	4·50	—	—
	39	51	29	40	20	13	1	3	1	—	—
1923	1·918	1·909	1·914	1·926	—	1·917	—	—	—	—	—
	0·45	1·13	1·55	2·00	—	3·00	—	—	—	—	—
	28	27	9	1	—	1	—	—	—	—	—

The meaning of each row is the same as that of the preceding tables. In Fig. 9, where these values are visualised, the curves for four years 1918–1921 are not much separated from one another, so that they can be combined into one, thus numerically:

Table XVIII.

Years	1·0 −1·4	1·5 −1·9	2·0 −2·4	2·5 −2·9	3·0 −3·4	3·5 −3·9	4·0 −4·4	4·5 −4·9	5·0 −5·4	<5·5
1918–21	1·943	1·948	1·949	1·943	1·946	1·949	1·947	1·947	1·946	1·950
	1·03	1·59	2·13	2·62	3·16	3·56	4·07	4·60	5·08	5·69
	46	36	74	56	87	42	53	64	117	61

These are shown in Fig. 10 together with the other data. Both in Fig. 9 and in Fig. 10, all the curves are practically horizontal, and only a general tendency of the solar constant to decrease along the year's advance, especially in later years, is apparent.

Now we come to a very definite conclusion: that is, there is no

direct relation practically between the solar constant and the calcium flocculi area, at least when they are *statistically* treated. The apparent regressions seen in Fig. 6 of the present Appendix and also in Fig. 10 of my October paper are illusory to some extent, being caused only by the distribution of weights. In other words, each of the curves shown in Figs. 7, 8, 9, and 10 is not a partial contribution to the general curves in Fig. 6; the natures of these two kinds of curves are quite different.

But, I do not mean by these conclusions to deny the possible relation between the two phenomena as obtained in the first part of my October paper. There they were treated *individually*, and the evidence is clear from actual examples.

My investigations, as a whole, include the results of two modes of attack on the problems of the solar constants: the short-lived variation and the long-enduring variation of the constant. As to the latter, the conclusion obtained just above is quite definitive. But, the method of treatment and the conclusions of the former part might be, in a sense, considered as not sound enough. As, however, this part was treated not statistically but individually, all the details were fairly complicated, so that the demontration of facts by means of some typical characters of actual phenomena was necessarily the only method possible. Moreover, the direct attack on these short-lived variations of the solar constant was the very point which Dr. Abbot induced me originally to investigate with our rich meterials. I do not think he expected me to use the statistical method of attack to explain his "four principles", notwithstanding my adopting this method in my last part. Each case of upheaval and depression of the solar constant of a few days' duration was no doubt expected to be explained by some corresponding apparitions on the solar disc. Under these circumstances, it may perhaps be necessary to preserve the main part of my October paper, especially those pages where the details of individual study of the solar surface from several sides, that is, from calcium K_2–plates and hydrogen C–plates are given as well as the data of magnetic polarity observations.

Therefore I know now that the variation of the heat radiation of the sun as received by the earth is not simple in its cause. Very rapid fluctuations within a week or so are probably explained by heliographic apparances when they are watched carefully by all the modern methods. But the general inclination of the solar radiation shown over a long period of several years, if any, is beyond our present understanding. I might say here that the decreasing mean latitude of solar-activity area towards the minimum epoch of the sun-spot may be, in some sense,

Fig. 6.

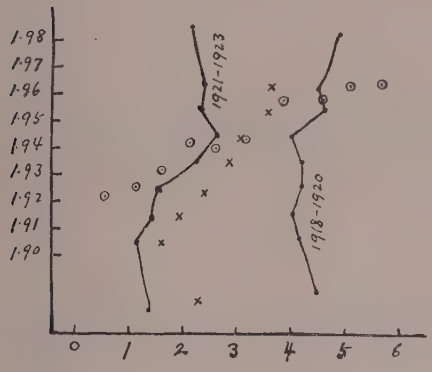


Fig. 7.

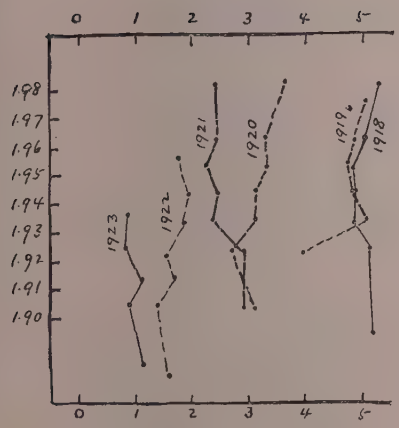


Fig. 8.

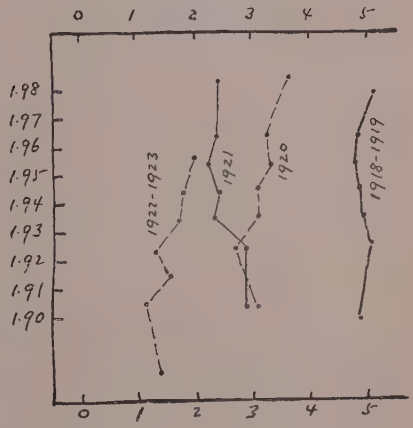


Fig. 9.

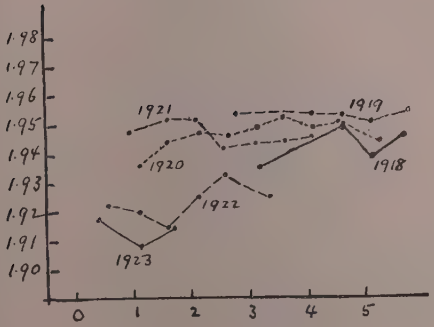
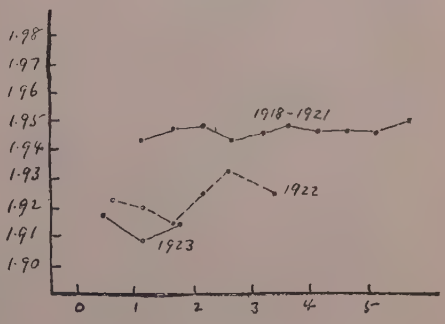


Fig. 10.



affecting the decided diminutions of the mean values of both solar constant and the flocculi area of the past few years.

Issei Yamamoto.

Harvard College Observatory,
Cambridge, Massachusetts, U. S. A.
February 28, 1924.

APPENDIX II.

On the Relation Between Sun-spots and the Solar Constants.[1]

Since 1921, we have a new series of sun-spot observations which are being made by Mr. Katué Misawa, Suwa, Nagano-ken. His programme is to observe sun-spots daily, making sketches of the solar disc with his 8-cm. telescope, visually, with a power of x83, and counting numbers of the spots and their groups. So that his works are quite similar to those of Zürich observers which are yielding the well known "relative numbers" of sun-spots for past several decades. The weather of Suwa, however, is much more uniform than that of Zürich according to Mr. Misawa's records of past years: and the distribution of numbers of days of solar observations all around the year is so uniform that a new series of "relative numbers" of sun-spots, computed by the writer recently, seems to give the more reliable data for the solar activity than from any other series.

Some weeks ago, the writer tried to find, if any, some relation between the sun-spot relative numbers from Misawa's observations and the series of observed solar constants from the Smithsonian Institution of Washington, D. C. The materials of the latter series are those given by C. G. Abbot, to the writer in 1923 during his staying at Mt. Wilson Observatory, and their dates of observations comprise the interval: 1918 July to 1923 May. Consequently the writer has taken up those materials in the present study which are from 1921 October to 1923 May, the interval being in common in both of these solar observations. All the investigations have been statistic, the treatments[2] of the materials being quite similar to that case in which the writer sought any relation between the areas of calcium flocculi and the solar constants in 1923.

In the first step, the whole material was divided into several groups according to definite intervals of the sun-spot relative numbers, and the mean values were obtained. The following table gives the results:

Table I.

Interval of Relative No.	0–9	10–19	20–29	30–39	40–49	50≦
Mean Value of Solar Const.	1·921	1·922	1·928	1·939	1·936	1·930
Mean Value of Relative No.	0·0	13·2	24·9	35·2	44·2	61·4
No. of Days	160	114	43	16	17	12

Similarly the whole materials were divided according to definite intervals of the solar constants, and mean values were obtained as tabulated below:

Table II.

Interval of Solar Const.	≦1·899	1·900 – 1·909	1·910 – 1·919	1·920 – 1·929	1·930 – 1·939	1·940 – 1·949	1·959 – 1·959	1·960≦
Mean Value of Solar Const.	1·884	1·906	1·914	1·925	1·934	1·944	1·955	1·965
Mean Value of Relative No.	6·8	8·7	12·5	11·1	14·6	18·1	16·7	17·4
No. of Days	37	42	71	61	62	49	18	21

Mean values of both of these two Tables have been plotted in Fig. 1, in which the small circles represent the values from Table I, and the crosses similarly those from Table II.

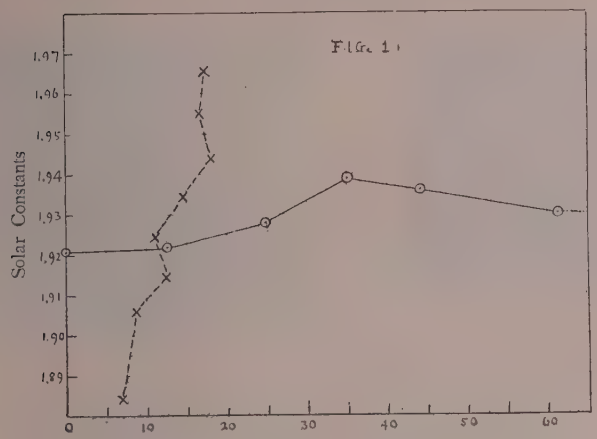
In Fig. 1, *some* kinds of correlations are suggested between the two series. To get a more definite results, the writer divided the whole interval of time into three parts, as:

1) 1921 October to 1922 March,
2) 1922 April to September,
3) 1922 October to 1923 May.

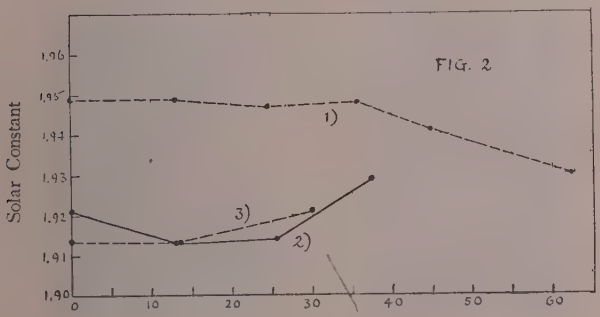
And, for each of these three intervals, the computations of obtaining mean values have been accomplished quite similarly as in the former cases. These results are summarised in Table III below:

Table III.

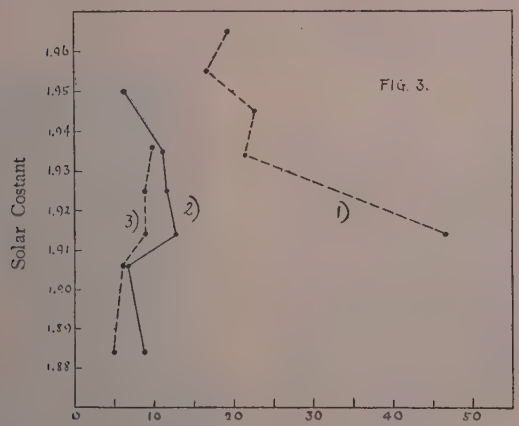
Epoch	0–9	10–19	20–29	30–39	40–49	50≦
1)	1·949	1·949	1·947	1·948	1·941	1·939
	0·0	13·0	24·5	35·7	44·8	62·3
	22	32	16	9	11	11
2)	1·921	1·913	1·914	1·929		——
	0·0	13·0	25·5	37·4		——
	59	37	15	10		0
3)	1·914	1·913	1·921			
	0·0	13·4	30·0			
	79	48	16			



Sun-Spot Relative Number from Misawa's Observations.



Sun Spot Relative Number from Misawa's Observations.



Sun-Spot Relative Number from Misawa's Obs.

For each epoch in the Table, the first row gives the mean solar constant, the second row the mean relative number, and the third row the number of days included.

Table IV.

Epoch	≦1·899	1·900 −1·909	1·910 −1·919	1·920 −1·929	1·930 −1·939	1·940 −1·949	1·950 −1·959	1·960≦
1)	——	1·914			1·934	1·945	1·955	1·965
	——	46·6			21·4	23·2	16·7	19·3
	0	8			22	33	18	19
2)	1·884	1·906	1·914	1·925	1·935	1·950		
	8·8	6·6	12·8	11·6	11·2	6·2		
	18	14	27	25	34	13		
3)	1·884	1·906	1·915	1·925	1·936	——	——	
	5·0	6·2	9·0	8·9	9·9	——	——	
	19	26	40	34	21	0	0	

The structure of this Table IV is quite similar to that of Table III. The results of these Tables are plotted in Fig. 2 and Fig. 3 respectively.

It is now clear that statistically speaking there is practically no relation between the sun-spot relative numbers and the solar constants, as seen in these Figures, in so far as our materials are concerned. This is the same conclusion as the writer obtained in 1923 for any apparent relation between the solar constant and the solar activity represented in the calcium flocculi on the disc.

Issei Yamamoto.

Kyoto University Observatory,
December 20, 1926.

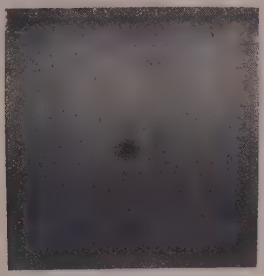
1 This paper is one of the contributions by the writer to the 3rd. Pan-Pacific Science Congress, held at Tokyo in 1926, and read at the Sectional Meeting on Nov. 2, 1926.

2 The daily values of the sun-spot relative numbers have been computed from the BULLETINs and THE HEAVENS of the Society of Astronomical Friends, Kyoto.

Plate IV.

Mt. Wilson Sun-spot No. 1884, on July 27, 1921, in *full development.*

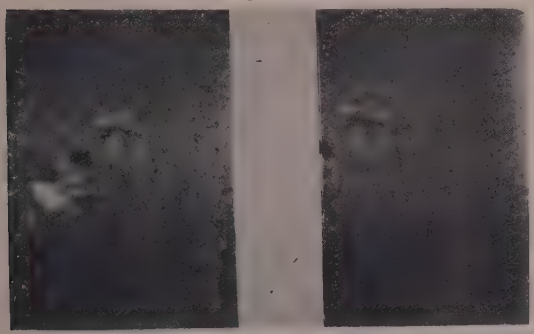
H*α* image.



Direct image.

Plate V.

Mt. Wilson Sun-spot No. 1904, on Sept. 19, 1921,
Heat effects just in *balance,*

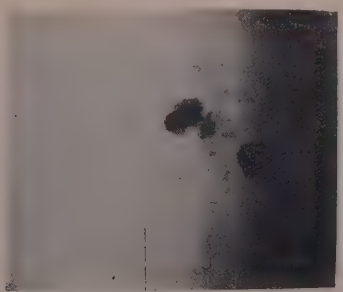


Both are H*α* images.

Plate VI.

Mt. Wilson Sun-spot No. 1842, on May 14, 1921; *decaying.*

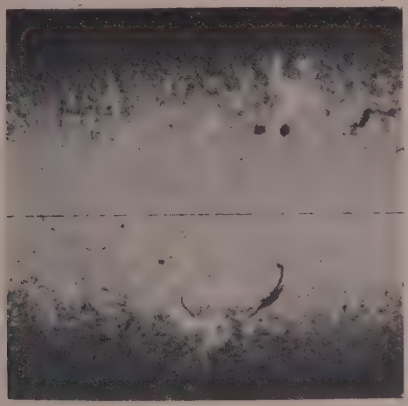
H*α* image.



Direct image.

Plate VII.

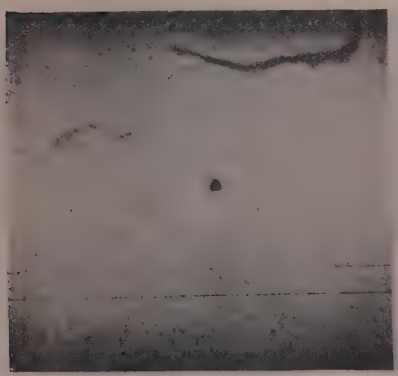
Mt. Wilson Sun-spot No. 1869, on July 7, 1921; in *decay*.



H*α* image.

Plate VIII.

Mt. Wilson Sun-spot No. 1917, on Nov. 18, 1921; *decaying*.

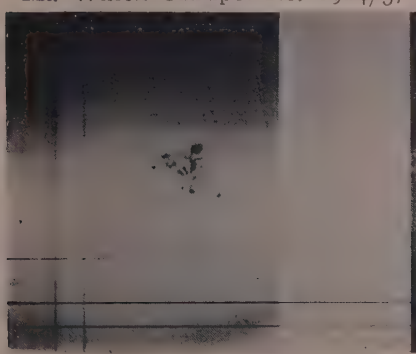


H*α* image.

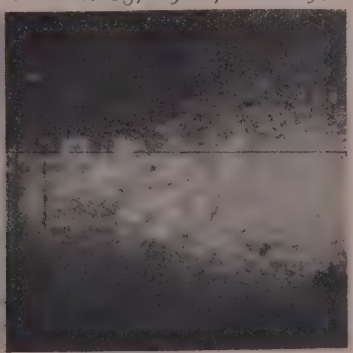
Direct image.

Plate IX.

Mt. Wilson Sun-spot No. 1924/5, on Dec. 15, 1921; in *decay*.



H*α* image.



Direct image.

A Peculiar Reversal of the Discharge-Figures on Photographic Plates

By

Usaburo Yoshida and Jinzo Tsutsumi

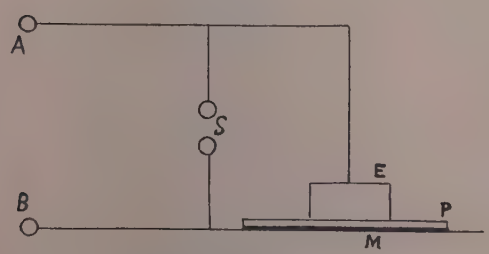
(Received April 23, 1928)

Abstract

A peculiar reversal of the discharge-figures on photographic plates, which is caused by fogging the plate with a spark-light simultaneously with the impression of the discharge-figures, is examined by the present experiment. It is found that the wave-length of the fogging light which is effective in this respect is shorter than about 420 μμ. It is also observed that this peculiar reversal is not due to a simultaneous illumination of the photographic plates with two lights of short duration.

About ten years ago, one of the writers[1] observed a peculiar reversal of the electric discharge-figures on a photographic plate, when the plate was illuminated by an electric spark simultaneously with the impression of the discharge-figures. Recently the same phenomenon was again examined; and though its final solution has not yet been attained it seems to have become a little clearer than before by the present experiment.

Fig. 1



The experimental arrangement is illustrated schematically by Fig. 1. An electrode E is placed on the sensitive film of a photographic plate P, which is covered beneath with a metallic plate M. These

1 U. Yoshida; These Memoirs, **2**, 315 (1917).

two electrodes E and M are connected to the two terminals A and B respectively, and a spark gap S is inserted in the manner represented by the figure. When the terminals A and B are connected momentaneously to both terminals of an influence machine respectively, a discharge–figure will be impressed on the photographic plate, and at the same time a spark will pass across the spark gaps, when its length is adjusted properly. Under these circumstances, the discharge–figure displays a peculiar reversal in the region illuminated by the spark, in addition to the effect of fogging by the light of the spark. When the discharge–figure is of the positive sign, every one of its irregular branches is enveloped by a somewhat diffuse margin of much stronger intensity as shown in Fig. 1, Plate I; and when the discharge–figure is of the negative sign, the general and regular outer boundary of its regular branches is enveloped by a somewhat diffuse margin of stronger intensity as shown in Fig. 2, Plate I.

It was stated in the former paper that the essential cause of this peculiar reversal was the simultaneity of the illumination of the spark and the impression of the discharge–figure; and consequently that, this being not an ordinary photographic reversal, nor the Clayden–effect, the presence of an intense electric field on the photographic plate at the momenent of formation of the discharge–figure may have an important bearing on this phenomenon. As to the effect of the illuminating spark, it was simply assumed in the former paper that this was due to the light emitted by the spark. This point was examined more closely in the present experiment by inserting various substances between the spark-gap and the photographic plate so as to screen the effect of the illuminating spark. The presence of a glass plate or a quartz plate as the screen does not essentially obstruct the occurrence of the peculiar reversal before mentioned, if we disregard the difference in the degree of the peculiar reversal, which is a little more weakened with a glass plate than with a quartz plate. This excludes the possibility of some corpuscular radiation being the agent from the illuminating spark to cause the peculiar reversal, as it will be entirely stopped by the glass plate or by the quartz plate. Next, it was observed, with the screen of a wire–gauze connected to earth, this screen did not offer any obstruction against the effect of the illuminating spark in any other region on the photographic plate than that of the shadow cast by the metal–wire of the gauze. This indicates that the effect of the illuminating spark in causing the peculiar reversal is not due to some electro–magnetic disturbance of comparatively long wave–length. These results seem to indicate

convincingly that the agent of the illuminating spark which is effective in causing the peculiar reversal of the discharge figure is the visible or ultra-violet light emitted by the spark. This consideration is further tested by screening off the light emitted by the spark with a sheet of black paper, and it was ascertained that a sheet of black paper is enough to stop the agent of the spark which is effective in producing the peculiar reversal. The next problem was to know the wave-length of the light which is emitted by the spark, and is effective in causing the peculiar reversal under consideration. This was examined by illuminating the photographic plate with the spark-light filtered by means of various colour-filters; and it was ascertained that the wave-length of the spark-light which was effective in causing the peculiar reversal was shorter than about 420 $\mu\mu$. This indicates that the light of the spark which is very effective in giving the photographic impression, is also very effective in causing the peculiar reversal under consideration. The effect of the polarization of the spark-light was also examined by letting the illuminating spark-light pass through a Nicol-prism at various orientations, and no detectable effect of the orientation of the plane of polarization of the illuminating spark-light upon the peculiar reversal was observed.

As to the part played by the discharge-figure on the photographic plate in giving rise to the peculiar reversal, a suggestion was given by one of the writers in the former paper that the strong electric field applied to the sensitive film at the moment of the impression of the discharge-figure has a complex rôle in some direct or indirect manner in causing the peculiar reversal. Another peculiarity of the discharge-figure on being impressed on the photographic plate was the momentariness of its light, by means of which its impression seems to be made on the photographic plate. If the peculiar reversal under consideration is due to the momentariness of the light of the discharge-figure, a simultaneous illumination of a photographic plate with two momentaneous lights will give rise to somewhat similar phenomenon as the peculiar reversal. This point was tested by an experiment illustrated by Fig. 2, and the result was negative. In Fig. 2, P is a photographic plate, S_1 and S_2 are two spark-gaps which are connected in parallel to the two terminals A and B. When these terminals are connected instantaneously to both terminals of an influence machine respectively, two sparks will pass across the spark gaps S_1 and S_2, and the photographic plate P will be illuminated simultaneously by the light emitted by these two sparks. By inserting in the position represented by Fig. 2 a screen C,

Fig. 2

which is provided with a narrow and long slit in a direction parallel to that of the spark gap S_2, the light emitted by the spark at S_2 is so constricted that only a narrow and long portion of the photographic plate is illuminated by it. On the other hand the light emitted by the spark at S_1 is made to illuminate a large portion of the photographic plate, so that a fogged image of the spark at S_2 is impressed on the photographic plate by simultaneous illumination. On about twenty photographs taken under such circumstances no trace of some similar phenomenon as the peculiar reversal under consideration was detected, so far as the present experiment is concerned. This indicates that the peculiar reversal is not due to a simultaneous illumination of the photographic plate with two instantaneous lights, and is in favour of the view that the presence of a strong electric field on the photographic plate at the moment of the impression of the discharge-figure is a primary cause in bringing about the peculiar reversal. The fact, mentioned before, that the fogging light of shorter wave-length is much more effective in causing the peculiar reversal seems to suggest that the peculiar reversal is due to the photo-electric effect of the fogging light upon the photographic plate which is subjected to a strong electric field.

Plate I

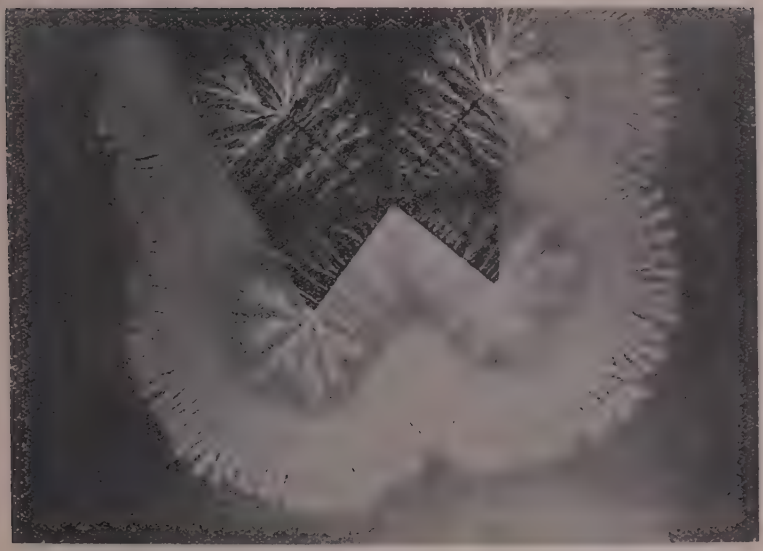


Fig. 1

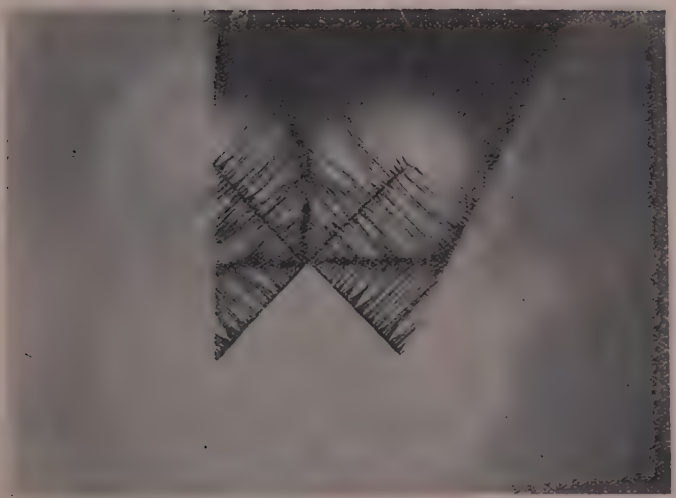


Fig. 2

On the Fibrous Structure in Metals Deposited by the Difference of Electrolytic Solutional Pressures

By

Shogo Tsuboi

(Received May 14, 1928)

Abstract

Metallic silver deposited by a solution of silver nitrate by putting a small piece of copper into it, has a fibrous structure, the common axis of which is 110 and makes an angle of 30° with the direction of the deposited siver. The micro-crystals show a rotation about this axis with an angle of some ±11°. As the (111) planes of the silver-crystal lie roughly parallel to the flat surfaces of the deposited metal, the direction of the growth of the deposited silver lies nearly parallel to the [112] axis.

Though the arrangement of the micro-crystals in electro-deposited metals have already been studied by many investigators, none of the metals deposited from their salt solutions by the difference of electrolytic solutional pressures have yet been examined. The writer has investigated, in the present experiment, the metallic silver which was deposited by a solution of silver nitrate owing to the presence of a piece of copper in it.

Specimen.

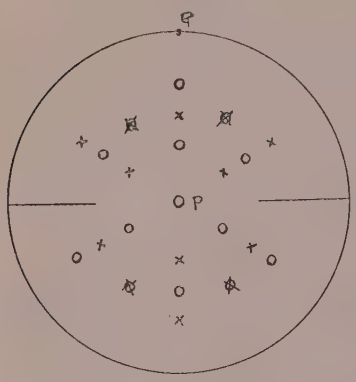
When we put a polished copper wire into a solution of silver nitrate the copper is covered, at the beginning, with black-coloured silver, which changes into grey with a further deposit, and after a few hours we can detect many long flat and acicular pieces of silver of the length of several mms. which are covered with smaller pieces branching off at an angle of about 60°.

The writer prepared many pieces of such deposited silver from the solution of the following three different concentrations: 0.3%, 0.6% and 2–4% in weight. We shall call the specimens obtained from these different solutions, Ag A_1, Ag A_2 and Ag A_3 respectively.

Experimental Results.

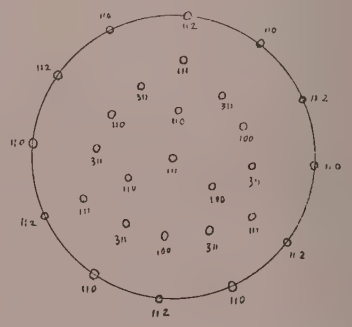
Ag A_1.—A specimen of the maximum breadth of 0.7 mms. and of a length of 38 mms. was obtained in this case. This specimen reflects light in many different directions as regularly as a single crystal of metal. If we project the directions of the normals of those reflecting planes in the stereographic projection, we get Fig. 1a. Where the pole P is the projection of the direction of the normal of the flat surface of the specimen, and the direction PG is parallel to the direction of the growth of the specimen (acicular axis).

Fig. 1a



⊠ is common to ○ and ×

Fig. 1b



On the other hand, the stereographic projection of the normals of various atomic planes of the cubic crystals, when one of its [111] axes is taken as the pole, is shown in Fig. 1b. Comparing these two figures, we know that Fig. 1a consists of two sets of points: the one represented by the small circles is identical with the same set in Fig. 1b, and the other, represented by the crosses, is the same as that obtained by rotating Fig. 1b through an angle of 180° about P. This fact seems to suggest that the surface normal of the flat surface of the specimen corresponds roughly to one of the [111] axes of the crystal, and the direction of the growth roughly to one of the [112] axes.

Under a microscopic examination, the specimen shows an aggrega-

tion of micro-crystals, with about 0.01 mms. in diameter. Every one has several faces which reflect light regularly in different directions.

Now the Laue-photographs were taken by using a molybdenum target. In doing so the direction of the growth of the specimen was set vertically, and the horizontal beam of the x-rays was made to strike the specimen in the direction perpendicular to the flat surface. Fig. 2, Plate I is the reproduction of the photograph thus obtained, and it consists of six principal radiating bands which make an angle of 60° with one other, and which are provided with the K spectra of molybdenum. The writer could easily find from this photograph that the common axis of the fibrous arrangement of the micro-crystals is the [110] axis of the crystal. The observed angles between the directions of the common axis (denoted by an arrow in Fig. 2 Plate I) and the lines drawn to the position of the K spectra on various radiating bands from the central spot are compared with the calculated values in Table I, and we see that the observed values agree fairly well with those calculated. The absence of several bands on the photograph seems to show that the fibrous structure corresponds to that of incomplete rotation. In order to estimate the angle of rotation of the micro-crystals, let us consider band IV in Fig. 2, Plate I ((111) plane, $\alpha = 90°$). If we consider the ideal arrangement of the micro-crystals without any rotation, as is suggested in the case of Fig. 1a, one of the (111) planes of the crystal is parallel to the surface of the specimen, and the other one of the (111) planes which is responsible to the band IV in Fig. 2, Plate I, makes an angle 19°30′ with the incident beam of the x-rays. On the other hand, for the same atomic plane of the crystal, the glancing angles for Mo K*a* are 8°40′ and 17°30′, respectively, for the first, and for the second order

Table I

Indices of the plane	$\alpha_{calc.}$	α_{obs} +	α_{obs} −	Band
311	27°	30°	30°	I
111	31°	30°	30°	I
111	34° 20′			
100	44° 10			
331	45° 20′	50°	48°	II
110	58° 50′			
311	63° 30′			
331	69° 30′	70°	70°	III
111	90°	90°	90°	IV
100	90°			
311	90°	90°	90°	IV
331	90°			
331	110° 30′	110°	110°	V
311	116° 30′			
110	121° 10′			
331	134° 40′	130°	132°	VI
100	135° 50′			
111	145° 40′			
111	149°	146°	152°	VII
311	153°	152°	149°	VII

spectra. So the reflection of the first order spectrum of K*α* of Mo will occur when the micro-crystals are rotated by the angle $19°30'-8°40'=10°50'$, from the ideal arrangement above mentioned. In all the photographs obtained by the writer, the first order spectrum of the K*α* line was just detected; and therefore it seems to be natural to estimate the angle of rotation of the micro-crystals about the axis of the fibre as $\pm 11°$.

If we draw the curves under the above considerations we get Fig. 2a. From the fact previously mentioned, in the case of Fig. 1a about the reflection of light, at last we reach Fig. 2b, in which are combined Fig. 2a, and the same one rotated through 180° in the plane of the paper. The general features of Fig. 2b are in fine accordance with Fig. 2, Plate I.

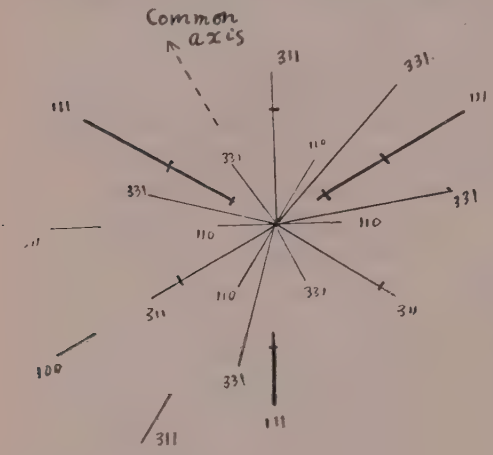


Fig. 2a

The above results may be stated in short, thus, that the micro-crystals have two ideal orientations as shown in Fig. 3, each one being obtained from the other by rotaing it through 180° in the plane of the paper. In these two ideal orientations, each one has one of the [111] axes in the direction parallel to the normal of the flat surface of the specimen, and one of the [112] axes in the direction of the growth; and the micro-crystals are arranged in a fibrous structure rotated through $\pm 11°$ about the common axis [110] from the two ideal orientations above mentioned.

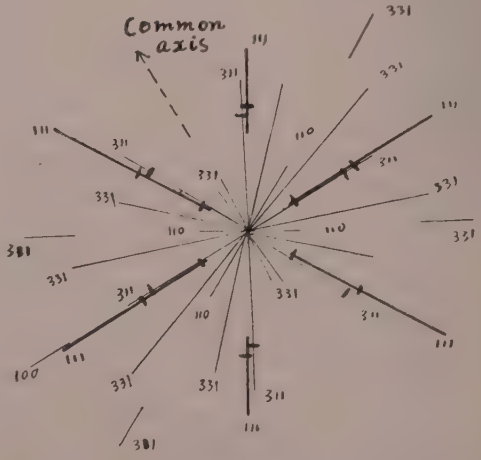


Fig. 2b

Ag A_2.—These speci-

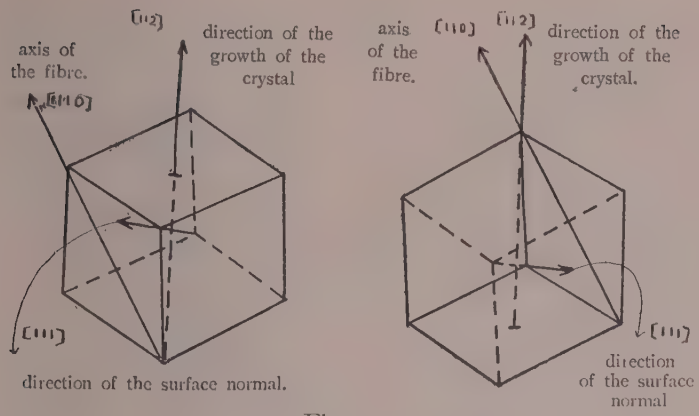


Fig. 3

mens were the same as Ag A_1 in light-reflection and in the Laue-photographs (Fig. 3, Plate I). In this case, however, having got good specimens branching off at 60°, Laue-photographs were taken of each branch; and it was seen that they were the same as Fig. 3 Plate I.

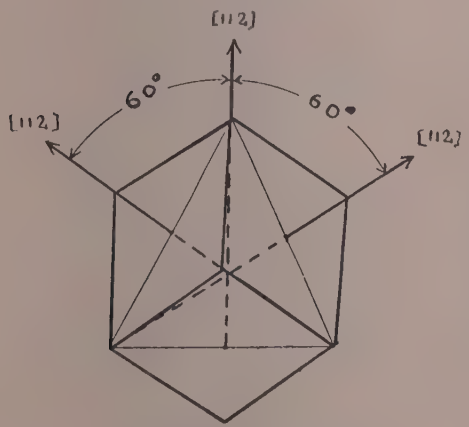


Fig. 4

From the fact that the direction of the growth is nearly parallel to the [112] axes, it seems to be understood why they branch out at 60°. Because these [112] axes lying in the same plane intersect at 60° to one other, and any one of these three axes may become the direction of the growth of the specimen.

*Ag A*3. —These were deposited from comparatively concentrated solutions after several days' standing, and they barely reflect light in many directions, as in the case of Ag A_1 and Ag A_2. A typical micro-photograph of the appearance of the specimen is shown in Fig. 4, Plate I. Taking the direction represented by the arrow in Fig. 5, Plate II, and one of the [110] axes of the crystal as the common axis of the fibrous arrangement, the position of the bands

Table II

Plane	α calc.	α + obs.	α − obs.	Band
311	27°	27°	——	I
111	31°	——	30°	I
111	34° 30′	——	——	
100	44° 10′	——	——	
331	45° 20′	46°	——	II
110	58° 50′	——	——	
311	63° 30′	——	——	
331	69° 30′	70°	——	III
111	90°	——	90°	IV
100	90°	——	——	
311	90°	90°	——	IV
331	90°	——	——	
331	110°	109°	——	V
311	116° 30′	——	——	
110	121° 10′	——	——	
331	131° 40′	132°	——	VI
100	135° 50′	——	——	
111	145° 40′	——	——	
111	149°	——	149°	VII
311	153°	151°	——	VII

is calculated under the consideration that there is only one of the two orientations shown in Fig. 3. The values calculated and observed are compared in Table II, and we can see a fair agreement. Sometimes we can obtain a very interesting specimen in this case, in which the rotation of the micro-crystals about the common axis becomes so small that we can treat it nearly as a piece of single crystal. One of the Laue-photographs taken of such a specimen is shown in Fig.6, Plate II; and from the distribution of the Laue-spots the writer was able to ascertain that the direction of the growth was in one of the [112] axes and the flat surface of the specimen was nearly parallel to one of the (111) planes of the crystal. This is in excellent accordance with the ideal orientation of the micro-crystals mentioned before.

The orientation of the micro-crystals thus determined with the aid of x-rays is in fine accordance with that suggested previously by examining the specimen from the side of the reflection of light. This result seems to indicate that the various atomic planes of the silver crystal the orientations of which are represented in Fig. 1b by the positions representing the direction of their surface-normals in stereographic projection, are developed actually as exterior surfaces of the silver-crystal.

Lastly, it must be noted that, for the testing of the fibrous structure and for the determination of the orientation of a crystal from the Laue-spots, the globe and the spherical scale constructed by Professor U. Yoshida[1] were mainly used.

1 U. Yoshida: Japanese J. Phys. **4**, 133(1927)

Summary.

(1) Metallic silver deposited by a solution of silver nitrate due to the presence of a piece of copper, consists of micro-crystals arranged fibrous-like in such a way that the common axis is the [110] axis of the silver crystal, and the micro-crystals are rotated about this axis by some $\pm 11^\circ$.

(2) The (111) plane of the crystal is situated nearly parallel to the flat surface of the specimen for the majority of micro-crystals.

(3) The axis of the fibre makes an angle of 30° with the direction of the growth of the specimen; therefore the direction of the growth is nearly parallel to the [112] axis.

The results of the present experiment agree well with that of H. Hirata and H. Kômatsubara's experiments[1] on electrolytic silver, excepting the point that the [112] axis, instead of the [110] axis in the case of electrolytic silver, is parallel to the acicular axis in the present case.

In conclusion, the writer wishes to express his sincere thanks to Professor U. Yoshida of Kyoto Imperial University for his kind guidance, and also to Mr. K. Yajima, the principal of Shiga Normal School, by whose kind permission the writer was able to obtain the necessary apparatus for this experiment.

Shiga Normal School

April 15, 1928.

1 These Memoirs, **10**, 95 (1926)

Plate I.

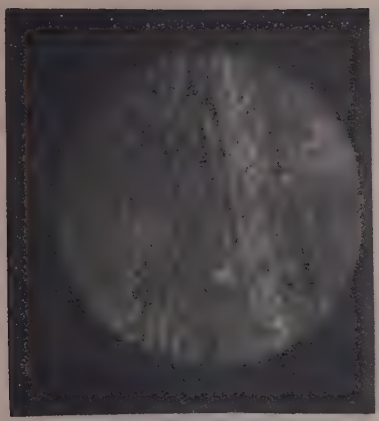


Fig. 1, Ag A_1
(magnification 100)

Fig. 2, Ag A_1

Fig. 3, Ag A_2

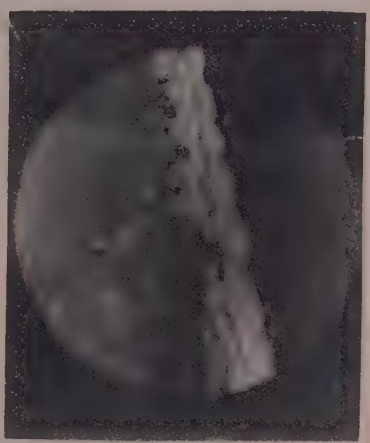


Fig. 4, Ag A_3
(magnification 100)

Plate II.

Fig. 5, Ag A_3

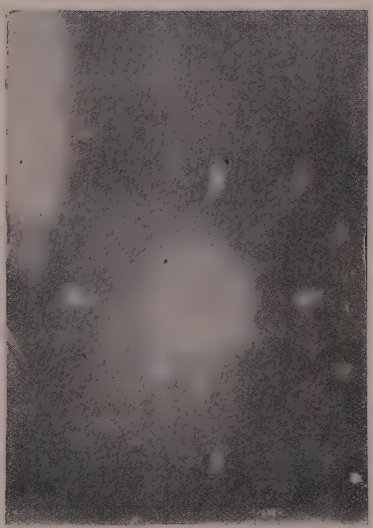


Fig. 6, Ag A_3

On the Destruction of the Single Crystal of Aluminium by the Process of Rolling.

By

Tokutaro Sakao.

(Received May 14, 1928)

Abstract

The scattering of the orientations of the micro-crystals, caused by rolling slightly the single crystal-plate of aluminium, was investigated in the present research. It was found that the micro-crystals rotated, up to some extent, around the axis parallel to the surface of the specimen and perpendicular to the direction of the advance of the process of rolling, irrespective of the orientations of the crystallographic axes referring to the surface of the specimen and the direction of the advance of the rolling process. The maximum angle of rotation of the micro-crystals was seen to increase proportionally with the reduction in thickness of the plate by rolling.

It is already known that, when a metal composed of a single crystal is subjected to some regular cold working, the single crystal is disintegrated into micro-crystals arranged in some regular way. Recently Prof. Sh. Tanaka[1] of the Kyoto Imperial University examined the arrangement of the micro-crystals of aluminium in the specimens prepared by rolling the single-crystal-plates of aluminium. But his experiments was mainly concerned with the specimens rolled thoroughly, and no detailed investigation of the arrangement of the micro-crystals was undertaken for the specimens rolled weakly and slightly. With a thin piece of a slightly rolled single crystal of a metal, the Laue-photograph taken by illuminating the specimen with a narrow pencil of x-rays is a little different from the ordinary Laue-spots; the ordinary sharp and well-defined Laue-spots become somewhat diffused and elongated to some extent, in this case, in some directions, the exent of this elongation and its direction being different

1 These Memoirs, **10**, 303 (1927)

with the degree of rolling and its direction. Such elongation of the Laue-spots must of course be due to some regular scattering of the orientations of the micro-crystals formed by the destruction of a single crystal-specimen in the process of rolling. At the suggestion of Professor U. Yoshida, the writer has tried to find from the elongation of the Laue-spots above mentioned the manner of such regular scattering of the orientations of the micro-crystals of aluminium.

The single-crystal-specimen in the form of a plate was prepared by the stress-annealing-method from a commercial aluminium plate of 1·5 mm. thickness. An ordinary Laue-photograph was taken with this single-crystal-plate. Then this single-crystal-plate was rolled successively with small steps of reduction in thickness, and at each successive stage of rolling a Laue-photograph was taken. The roller used was of two steel cylinders of 15 cms. in diameter. As the single-crystal-plate was very liable to bend in an irregular manner by rolling, it was rolled by being held between two brass plates of 0·5 mms. thickness; and by this means the irregular bending of the specimen caused by the rolling was mostly avoided. As to the direction of rolling, special care was taken to roll always in the same direction for each specimen.

The orientation of the crystallographic axes of the undeformed initial single crystal of aluminium, in reference to the outside face of this single-crystal-plate, was determined by dealing with the Laue-spots for this single-crystal-plate by means of the spherical scale and the globe devised by Prof. U. Yoshida.[1] When the single-crystal-plate is rolled a little, each one of the ordinary sharp and well defined Laue-spots become elongated to some extent in some direction, the extent of this elongation and its direction being different for different spots. As was stated before, this elongation of the Laue-spots must be due to some regular scattering of the orientations of the micro-crystals of aluminium which is caused by rolling the single-crystal-plate of aluminium. If so, both ends of every elongated Laue-spot must correspond to two extremities of the scattered orientations of the micro-crystals; and the whole assemblage of the points representing the positions of both ends of every elongated Laue-spot on the photographic plate will be two sets of Laue-spots due to the micro-crystals at the two extreme orientations above mentioned. As an intermediate portion between the two ends of every elongated Laue-spot is caused by the micro-crystals at a certain intermediate orientation between the two extreme ones, the problem will be solved if we can determine

1 Jap. J. phys., **4**, 133 (1927)

these two extreme orientations from the elongated Laue-spots. Actually these two extreme orientations of the micro-crystals were found easily with the aid of the spherical scale and the globe before mentioned.

Thus, for the present purpose, it is very important to know as accurately as possible the positions of both ends of an elongated Laue-spot on a photographic plate. For this purpose the writer employed a Coolidge-x-ray-tube having a tungsten target. With the x-ray-tube having a target of another metal, such as molybdenum, the intensity of the characteristic radiation of that metal was much stronger than that of the continuous radiation, so that the appearance of the spectrum of the strong characteristic radiation on the continuous background in an elongated Laue-spot prevented sometimes a reliable determination of the positions of the end-points of the elongated Laue-spot. However with the tube having a tungsten target, the intensity of the impression of the elongated Laue-spots became rather uniform over the whole part of every spot, which was of course due to a rather uniform distribution of the intensities of the continuous x-rays for various wave-lengths. This fact enabled the writer, by the use of the tube with tungsten target, to determine definitely the positions of the end-points of the elongated Laue-spots.

In taking the Laue-photograph, a narrow and circular beam of the x-rays was made to illuminate the aluminium plate in a direction perpendicular to its flat surface, and the photographic plate was placed behind the specimen at a certain distance. This distance between the specimen and the photographic plate was kept the same throughout the present experiment, so that the continuous change of the Laue-photographs with the lapse of the stages of rolling could be easily traced by overlapping the photographs.

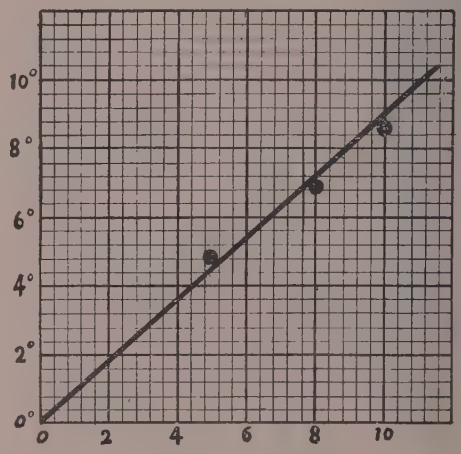
The Laue-photographs taken are reproduced in Figs. 1, 2, 3, 4 and 5, Plate I. Fig. 1 is an ordinary Laue-spots taken with a single-crystal-plate of aluminium, and the remaining four figures are taken with the same specimen at its four successive stages of rolling. The reductions in thickness of the plate by the rolling were 2, 5, 8 and 10 percent of the original thickness respectively, corresponding to Figs. 2, 3, 4 and 5, Plate I. In all the photographs in Plates I, the horizontal direction passing through the centre of the direct image is parallel to the flat surface of the specimen, and is perpendicular to the direction of the advance of the rolling process on the surface of the specimen.

From the figures in Plate I we know that the elongation of each Laue-spot increase with the increase of the reduction in thickness of the plate by rolling, and that for different spots in the same figure this

elongation is much smaller with the spot which subtends a smaller angle at the centre of the direct image with the horizontal direction passing through the centre of the direct image on the surface of the figure. This fact seems to suggest that the scattering of the orientations of the micro-crystals caused by the process of rolling is such that the micro-crystals rotate, up to some extent, around the axis parallel to the surface of the specimen and perpendicular to the direction of the advance of the process of rolling. This suggestion was confirmed to be so by the aid of the spherical scale and the globe, and the maximum angle of rotation of the micro-crystals around the said axis was measured on the spherical scale. The relation between this maximum angle of rotation of the micro-crystals and the reduction in thickness of the plate by rolling is shown in Fig. 1, and the proportionality between these two quantities is observed to hold nearly so far as the present experiment is concerned.

The writer has repeated the same experiment with seven single-crystal-plates of aluminium of different orientations of the crystallographic axes, and found that the scattering of the orientations of the micro-crystals by the process of rolling was the same as that stated above irrespective of the orientation of the crystallographic axes referring to the surface of the plate and the direction of the progress of the rolling process.

Fig. 1.



It will be very interesting to see whether or not the scattering of the orientation of the micro-crystals proceeds in the manner above stated, when the degree of rolling is very much more increased; and the experiments in this direction are now going on.

In conclusion, the writer wishes to express his deepest thanks to Professor U. Yoshida of the Kyoto Imperial University, under whose guidance the present research was completed.

Plate I.

Fig. 1.

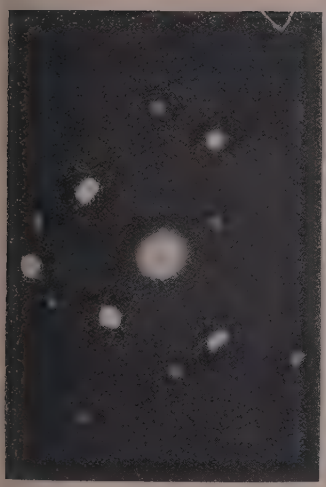


Fig. 2.

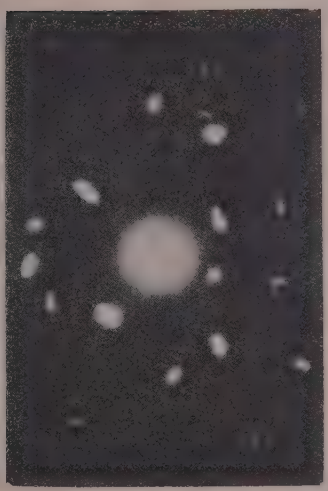


Fig. 3.

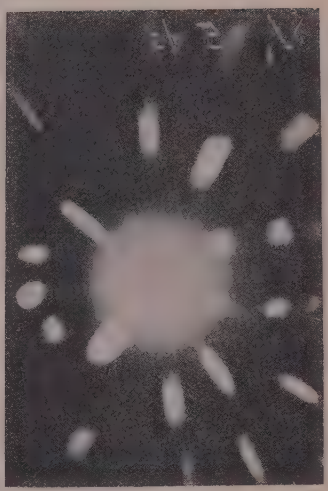


Fig. 4.

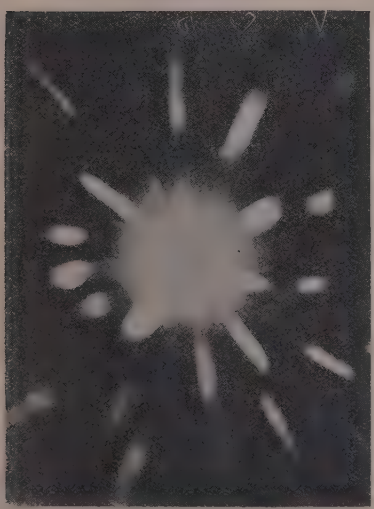
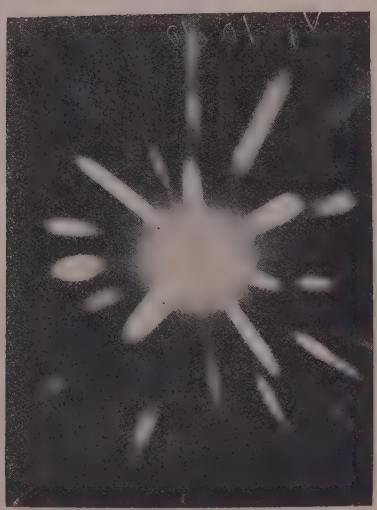


Fig. 5.



Laue-Photograph taken with Convergent X-Rays

By

Takeo Fujiwara.

(Received May 23, 1928)

Adstract

Some time ago, the writer had determined the orientation of the crystals in tungsten wire by finding the spectrum of the characteristic K-radiation of the Mo-target in some of the ordinary Laue-spots. Now he has improved some parts of the former method. He utilized the convergent X-rays starting from the focus of the Mo-target of a Coolidge tube. These convergent X-rays were made to pass through a circular pin hole, and then to illuminate a small portion of a specimen. The photographic plate was set perpendicular to the line connecting the centres of the focus and of the slit. In all photographs taken with convergent X-rays, we found that each of many Laue-spots contained an intense nucleus at its centre, and that some of the spots were accompanied by the spectrum lines of the K-radiation of molybdenum. As to the positions of the spectrum lines, they did not, generally coincide with those of the corresponding centres of the Laue-spots. From the positions of these spectrum lines and of the intense centres the orientation of the crystal was determined. In obtaining the exact position of the centre of the central spot, which was impressed by the direct beam of the X-rays, two methods were employed. One was to give a correction, by calculation, of its rough position estimated provisionarily; and the other was to use crossed wires before the photographic plate.

Introduction

Some time ago, the writer[1] had determined the orientation of the crystals in tungsten wire by finding the characteristic K-radiation in some of the ordinary Laue-spots.

1 T. Fujiwara, Mazda-Kenkyu-Jiho, **1**, No. 2, (1926)

At that time the X-rays passing through the slits of two pin-holes were used, the specimen and the photographic plate were generally placed perpendicular to the beam of the X-rays. Recently the same method has been improved in certain particulars by the writer and is described in the following pages.

General Idea of using Convergent X-Rays

If a crystal be placed in a position satisfying the following Bragg's equation in the path of monochromatic X-rays of wave length λ, it will reflect them according to

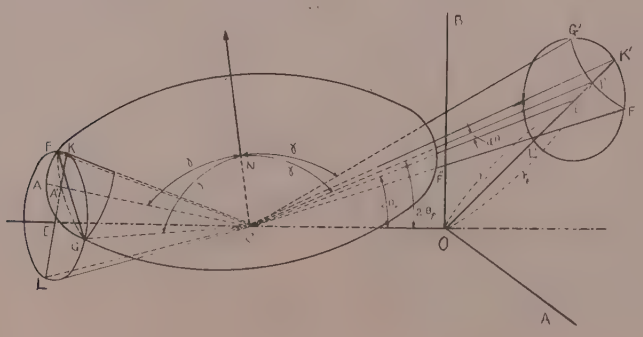
$$n\lambda = 2d \sin\theta, \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

where d is the grating constant of the crystal, and θ is the angle between the atomic plane of the crystal and the incident ray, and n is an integer which determines the order of the spectrum. But if a crystal be placed in the path of the "white" X-rays, it will reflect some of them at any position of the atomic plane, as there will be some X-rays of a certain wave length in the incident X-rays, which will satisfy equation (1). The diffraction pattern thus taken is so-called Laue-spots.

Therefore if the "White" X-rays containing a characteristic radiation of strong intensity strike the crystal plane in such a position that the wave length of the characteristic radiation satisfies equation (1), the spectrum line of that radiation will be found in the ordinary Laue-spots.

Now let convergent "white" X-rays accompanying a characteristic radiation of strong intensity strike a crystal plane and produce a Laue diffraction pattern on the photographic plate in the manner shown in Fig. 1. In this figure CKL is convergent rays starting from a

Fig. 1



circular focus KFLG, E is the centre of it, and the plane POB is the photographic plate which is placed perpendicularly to the line EC.

Now we assume any characteristic radiation AC which is contained in the plane ECN and is reflected by an atomic plane of the crystal in the direction CP. CN is the normal to the atomic plane, γ the angle between the ray and the normal to the plane, and P the image on the photographic plate which is impressed by the reflected characteristic radiation. Then, in addition to the ordinary Laue-spot, the same characteristic radiation which strikes the crystal at the same glancing angle as the former one, will also be reflected by the same atomic plane, and the characteristic radiations whose paths are the generators of the cone CFAGF'' will be reflected by the plane and will make a spectrum line G'PF' in the ordinary Laue-spot. The vertex of the cone CFAGF'' is C, its axis CN and the semivertical angle γ.

The characteristic radiations whose paths are not lying on the surface of the cone CFAGF'', do not satisfy equation (1), they will not be reflected by the same atomic plane. Thus, the shape of the spectrum line produced on the photographic plate will be, strictly speaking, a curved line G'PF', but as the angle γ is very near to $\frac{\pi}{2}$, so the spectrum line appears nearly as a straight line and is perpendicular to the radial line OP.

Compared with the case of using parallel rays, the sharpness of the Laue-spots is of course very much destroyed by using convergent X-rays; but the chance of obtaining the spectrum lines in the Laue-spots is very much increased with convergent X-rays, because the chance of covering the focus by the conical surface CFAGF'' is greatly increased by using a broad focus tube.

Outline of the Method

As stated above the writer has improved some parts of the former method. He utilized convergent X-rays starting from the focus on the Mo-target of a Coolidge tube. These convergent X-rays were made to pass through a circular pin hole, and then to illuminate a small portion of a specimen of a single crystal of tungsten. The photographic plate was set perpendicular to the line connecting the centres of the focus and of the slit. For the present purpose an X-ray tube with a broad focus and a circular conical slit are preferable. The focus of the tube was about 9 mm in diameter and the applied voltage was about 40 K.V.s. The semivertical angle of the conical slit was about five degrees, slightly

larger than that of the convergent X-rays, and the diameter of its opening was about 0.4 mm.

A lead screen with a circular hole of proper diameter was placed between the X-ray tube and the slit so as to cut off the weak X-rays starting from any other part of the anode than the focus. By the use of this lead screen the Laue-spots and the central spot impressed by the direct beam of the X-rays became sharper and thinner than those obtained without a screen.

In setting the photographic plate perpendicularly to the line joining the centres of the focus and of the slit, a fluorescent screen provided with a point-mark was so placed, just at the position where the photographic plate would be placed afterward, that the line connecting the centre of the slit and the point-mark became perpendicular to the plane of the fluorescent screen. Then the position of the X-ray tube was so adjusted as to project the image of the centre of the focus at the point-mark on the screen: and then it was replaced by the photographic plate exactly in the same position.

The Results

With this apparatus Laue diffraction patterns of thin tungsten wires composed of single crystals were taken as shown in Figs. 1 & 3 of Plate I, and the diffraction pattern of the same crystal taken with the parallel X-rays is shown in Fig. 2 of Plate I. As comparison, the Laue patterns in Figs. 1 & 2 were taken with the same tungsten crystal in the form of a thin wire, which was exactly in the same position. The potential difference applied on the X-ray tube was about 40 K.V.s, the current about 10 milliamperes and the exposure of the photographic plates about five hours in both cases.

Comparing these two photographs we can see that some more spectrum lines of the characteristic K-radiation of Mo appeared in Fig. 1, and that the lines are longer in this case. With regard to the Laue-spots produced by the white radiation they are larger in Fig. 1 than in Fig. 2.

By means of convergent X-rays and with a crystal of tungsten the writer took many photographs with different X-ray tubes having foci of different shapes; and it was observed, as a matter of course, that the shapes of the Laue-spots were respectively similar to those of the corresponding central spots which are nothing but the ordinary images of the foci obtained by the pin hole slit. This is illustrated by

the photographs, shown in Figs. 1, 3, 4, 5, 6, 7, & 8 of Plates I & II. In the cases of Figs. 1, 3 & 4, Plate I, the same anode with a nearly circular focus was used, the shape of which is shown in Fig. 7, Plate II. With Fig. 4, Plate I and Fig. 5, Plate II, the same tungsten crystal was used, but with the others the tungsten crystals used were different in each instance.

In all photographs taken with convergent X-rays we can detect that every one of the Laue-spots has an intense nucleus at the centre. This is, of course, due to the presence of an intense nucleus at the centre of the focus of the target; and it is very convenient in determining the position of the Laue-spot, and consequently in determining the orientation of the crystal. As to the position of the spectrum lines of the K-radiation, they are not generally coincident with the corresponding intense nuclei at the centres of the Laue-spots. The glancing angle θ of the X-rays which are responsible for the spectrum line G'PF' in Fig. 1 does not coincide with the half of the angle PCO, but it will be given by the following equation.

$$2\theta = 2\theta_n \pm 2d\theta, \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (2)$$

were θ_n is the glancing angle of the intense nucleus at the centre of the Laue-spot E', $d\theta$ is the difference between the angles $PCO = 2\theta_p$ and $E'CO = 2\theta_n$, and the double signs are to be properly selected according to the position of the spectrum line against the point E'. This circumstance must be taken into consideration in determining the orientation of the crystal.

The determination of the orientation of the axes of the crystal with these spectra and spots will be carried out as follows:

At first, from the measured values of r_n and r_p which are the distances between OE' and OP respectively, the values of the angles E'CO, and PCO and their difference $d\theta$ are calculated. The glancing angle θ corresponding to the spectrum line is found by equation (2), and the indices of the atomic plane of the crystal by whose reflection the spectrum line and Laue-spot are caused, are determined in the same way as in the former method. From the value of θ and the observed value of the angle between the lines OE' and OB which is drawn on the photographic plate in the direction parallel to the axis of the wire, the orientation of the crystal is determined in the ordinary manner.

Table I

No. of spots.		r (obs.) in mm.	$2\theta_n$ or $2\theta_p$ in degree		$d\theta$ in degree	θ (calc.) in degree	Grating constant d in 10^{-8} cm.		Davey's Data d in 10^{-8} cm.	Davey's Data Indices of atomic plane	Indices of the atomic plane.
	r_n	18·19	θ_n	31°39.′5							
							$d\alpha'$	1·56	1·58	(100)	
							$d\alpha$	1·54	1·58	(100)	
	r_{p1}	16·39	θ_{p1}	29°3.′4	−2°36.′1	13°13.′65	$d\beta$	1·38			$(100)_\alpha$
1							$d\gamma$	1·35			
							$d\alpha'$	1·73			
							$d\alpha$	1·72			
	r_{p2}	15·46	θ_{p2}	27°42.′3	−3°57.′2	11°52.′6	$d\beta$	1·53	1·58	(100)	$(100)_\beta$
							$d\gamma$	1·51			
	r_n	27·55	θ_n	43°2.′6							
							$d\alpha'$	0·853	0·843	(321)	
							$d\alpha$	0·848	0·843	(321)	
	r_{p1}	30·76	θ_{p1}	46°11.′9	+3°9.′3	24°40.′6	$d\beta$	0·756			$(321)_\alpha$
2							$d\gamma$	0·742			
							$d\alpha'$	0·964			
							$d\alpha$	0·958			
	r_{p2}	27·77	θ_{p2}	43°16.′1	+ 13.′5	21°39.′8	$d\beta$	0·855	0·843	(321)	$(321)_\beta$
							$d\gamma$	0·839			
	r_n	29·00	θ_n	44°30.′6							
							$d\alpha'$	1·14	1·12	$(110)^2$	$(110)^2_\alpha$
3	r_p	25·21	θ_p	40°31′	−3°59.′6	18°15.′65	$d\alpha$	1·13	1·12	$(110)^2$	or
							$d\beta$	1·01	0·997	(310)	$(310)_\beta$
							$d\gamma$	0·989	0·997	(310)	

Table I (cont.)

No. of spots.		r (obs.) in mm.	$2\theta_n$ or $2\theta_p$ in degree		$d\theta$ in degree	θ (calc.) in degree	Grating constant d in 10^{-8} cm.		Davey's Data d in 10^{-8} cm.	Davey's Data Indices of atomic Plane	Indices of the atomic plane
	r_n	25·18	θ_n	40°29′							—
							$d\alpha'$	0·980			
							$d\alpha$	0·974			
	r_{p1}	26·15	θ_p	41°33′3	+1°4′3	21°18′8	$d\beta$	0·862			$(321)_1$
4							$d\gamma$	0·853	0 843	(321)	
							$d\alpha'$	0·961			
							$d\alpha$	0·956			
	r_{p2}	26·54	θ_p	41°58′55	+1°29′35	21°44′05	$d\beta$	0·852	0·843	(321)	$(321)_3$
							$d\gamma$	0·837			
	r_n	21·28	θ_n	53°48′3							
							$d\alpha'$	1·31	1·29	(211)	$(211)_\alpha$
	r_{p1}	19·63	θ_p	33°38′5	−2°9′8	15°44′35	$d\alpha$	1·31	1·29	(211)	
							$d\beta$	1·16			or $(110)^2_\gamma$
5							$d\gamma$	1·14	1·12	$(110)^2$	
							$d\alpha'$	1·48			
							$d\alpha$	1·47			
	r_{p2}	18·29	θ_p	31°48′	−4°0′3	13°53′85	$d\beta$	1·31	1·29	(211)	$(211)_3$
							$d\gamma$	1·29	1·29	(211)	
	r_n	9·55	θ_n	17°56′2							
							$d\alpha'$	2·28	2·23	(110)	
6	r_p	9·55	θ_p	17°56′2	0	8°58′1	$d\alpha$	2·27	2·23	(110)	$(110)_\alpha$
							$d\beta$	2·02			
							$d\gamma$	1·99			

From the diffraction patterns shown in Fig. 3, Plate I, the writer determined the indices of the atomic planes by whose reflections the spectrum lines and the Laue-spots are caused, and are given in Table I. In this table the spots 1, 2, 3, etc., refer respectively to those marked in Fig. 3, Plate I. The values of r_p and r_n corresponding to the spectrum lines and the intense nuclei at the centres of the Laue-spots respectively were measured on the negative plate with Hilger's travelling micro-meter and are represented in the 3rd column. For the value of the distance between the specimen and the photographic plate a=29.50 mms. was used. The angles $2\theta_n$ and $2\theta_r$ in the 4th column correspond respectively to the intense nucleus and the spectrum line in the same horizontal row. The symbols $d_{\alpha'}$, d_α, d_β & d_τ in the 8th column represent the value of d obtained by assuming that the spectrum tabulated in the same horizontal row is due to any one of the $K_{\alpha'}$, K_α, K_β and K_τ lines of molybdenum respectively. For the value of the wave length of the $K_{\alpha'}$, K_α, K_β and K_τ lines the writer has taken $\lambda_{\alpha'}=0.712\times10^{-8}$ cm., $\lambda_\alpha=0.708\times10^{-8}$ cm., $\lambda_\beta=0.631\times10^{-8}$ cm. and $\lambda_\tau=0.620\times10^{-8}$ cm.[1] respectively. In the 10th and the 11th columns, the values of d and the indices of the corresponding reflecting atomic plane obtained by Davey[2] are represented. The symbols $(100)_\alpha$, $(100)_\beta$, etc., in the last column show that the corresponding line is the spectrum of the K_α or K_β line caused by the reflection from the atomic plane represented by the indices given in the bracket, and the symbol $(110)^2$ denotes that the spectrum line is the second order spectrum caused by the crystal.

Correction for the Determination of the Position of the Centre of the Central Spot

In the former case the photographic plate was set perpendicular to the line connecting the centres of the nucleus of the focus and of the slit exactly, and the distance between the photographic plate and the specimen was accurately measured. The centre of the central spot, which was impressed by the direct beam of the X-rays, was determined on the negative plate by drawing two diametral lines on the central spot. The shape of this central spot was nearly a circle and could clearly be seen on the original plate, though it was barely visible in the reprinted figure.

Generally the centre thus obtained does not coincide exactly with

1 W. Duane and Kang-Fuh-Hu: Phys. Rev., **9**, 489 (1918); **14**, 369 (1919).

2 W.P Davey: Phys. Rev., **25**, 678 (1925).

the intense nucleus of the central spot, and in the former experiment its accuracy was much less than the other factors stated above. Therefore the writer obtained the position of the centre of the central spot, which is more close to the real one than that obtained in the former, by making a correction. But if we are doubtful of the accuracies of the distance between the photographic plate and the specimen, and of the setting of the photographic plate perpendicularly to the line connecting the centres of the nucleus of the focus and of the slit, besides the correction for the centre of the central spot, as a matter of course, we must consider the corrections for these factors. The correction for the centre of the central spot was made as follows:

Let the real centre of the intense nucleus of the central spot which is impressed by the direct beam of X-rays on the photograph be O, its approximate position be O′, the centre of the intense nucleus of the reflected Laue-spot be E′, and the reflected spectrum line of a known wave length be P′P as shown in Fig. 2. In this figure all symbols have the same meaning as in the case of Fig. 1.

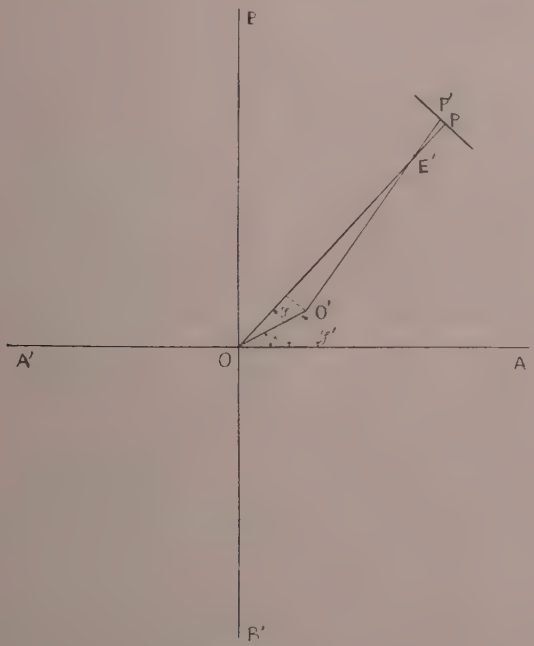


Fig. 2

If we put $OO'=c$, angle $O'OA=\alpha$, $OE'=r_n$, $OP=r_p$, $O'E'=r'_n$ and $O'P'=r'_p$, then we have the following relations

$$\left.\begin{aligned} \tan 2\theta_n &= \frac{r_n}{a} \\ \tan 2\theta_p &= \frac{r_p}{a} \\ \tan 2\theta'_n &= \frac{r'_n}{a} \\ \tan 2\theta'_p &= \frac{r'_p}{a} \end{aligned}\right\} \quad \cdots\cdots (3)$$

$$\left.\begin{aligned} r_n &\fallingdotseq r'_n + c\cos(\varphi'-\alpha) \\ r_p &\fallingdotseq r'_p + c\cos(\varphi'-\alpha), \end{aligned}\right\} \quad \cdots\cdots (4)$$

where θ'_n and θ'_p are the values corresponding to the centre O', the symbols without dash represent the values corresponding to the real centre and φ' is the angle between the lines E'O' and OA.

From the equations (3) and (4) we have

$$\tan 2\theta_n = \frac{r'_n}{a} + \frac{c}{a}\cos(\varphi'-\alpha),$$

$$\text{i. e. } \tan 2\theta_n - \tan 2\theta'_n = \frac{c}{a}\cos(\varphi'-\alpha). \quad \cdots\cdots (5)$$

Now, at first, take the approximate position of the centre of the central spot as O', and after finding the values θ'_n and $d\theta'$, determine the indices of the atomic plane by whose reflection the spectrum line P'P is caused in the manner as was stated before. Then by using Davey's data, calculate the glancing angle θ of the above determined spectrum line by equation (1). With this value of θ and that of $d\theta'$ which was obtained before, the value of θ_n corresponding to that spectrum line is calculated by the following equation

$$\theta_n = \theta \mp d\theta' \quad \cdots\cdots (6);$$

and then the value of $\tan 2\theta_n$ will be immediately obtained.

Generally the difference $\tan 2\theta_n - \tan 2\theta'_n$ does not vanish and it takes the value $\frac{c}{a}\cos(\varphi'-\alpha)$.

If we could find the values of c and α which satisfy equation (5) for the various spots, we can obtain the corrected values of r_n and r_p, and then the real value of θ of the spectrum line.

For the purpose of obtaining the values c and α, at first the relations between φ' and $\tan 2\theta_n - \tan 2\theta'_n$ for various spots are required; and then by taking the value of $\tan 2\theta_n - \tan 2\theta'_n$ as the radius vector and φ' as the vectorial angle these relations are plotted for various spots. If a circle passing approximately through the origin and the plotted points is drawn, and the length of the diameter of this circle which passes through the origin and the vectorial angle of this diameter are taken as $\frac{c}{a}$ and α respectively, then these values of $\frac{c}{a}$ and α will approximately satisfy equation (5). Thus the approximate values of c and α which satisfy equation (5) can be obtained.

If we denote these approximate values of c and α by c_0 and α_0 respectively, the real values of c and α will be represented as follows

$$c = c_0 + \Delta c \qquad \alpha = \alpha_0 + \Delta\alpha, \quad \cdots\cdots\cdots\cdots\cdots\cdots (7)$$

where Δc and $\Delta\alpha$ are very small.

Substituting these relations into equation (5) the following relations are derived,

$$\tan 2\theta_n - \tan 2\theta_n' = \frac{c_0 + \Delta c}{a} \cos(\varphi' - \alpha_0 - \Delta\alpha)$$

$$= \frac{c_0 + \Delta c}{a} \cos(\varphi' - \alpha_0) \cos(\Delta\alpha) + \frac{c_0 + \Delta c}{a} \sin(\varphi' - \alpha_0) \sin(\Delta\alpha).$$

By putting $\cos(\Delta\alpha) = 1$, and $\sin(\Delta\alpha) = \Delta\alpha$ the above equation becomes

$$\tan 2\theta_n - \tan 2\theta_n' = \frac{c_0}{a} \cos(\varphi' - \alpha_0) + \frac{\Delta c}{a} \cos(\varphi' - \alpha_0)$$

$$+ \frac{c_0 + \Delta c}{a} (\Delta\alpha) \sin(\varphi' - \alpha_0). \quad \cdots\cdots\cdots\cdots\cdots (8)$$

Since α_0 and c_0 are known, the values of $\frac{c_0}{a} \cos(\varphi' - \alpha_0)$ and $(\varphi' - \alpha_0)$ can be calculated for all points. By putting

$$\left\{ \tan 2\theta_n - \tan 2\theta_n' \right\} - \frac{c_0}{a} \cos(\varphi' - \alpha_0) = \Delta \quad \cdots\cdots\cdots\cdots\cdots (9)$$

the following relation is obtained from equation (8)

$$\frac{\Delta}{\cos(\varphi' - \alpha_0)} = \frac{\Delta c}{a} + \frac{c_0}{a} (\Delta\alpha) \tan(\varphi' - \alpha_0). \quad \cdots\cdots\cdots\cdots\cdots (10)$$

By putting

$$\frac{\Delta}{\cos(\varphi'-\alpha_0)}=y \quad \text{and} \quad \tan(\varphi'-\alpha_0)=x,$$

the values of x and y are calculated for all spots, and if the relation between x and y is plotted on a graph it will be represented nearly by a straight line. By drawing this line properly the values of $\frac{\Delta c}{a}$ and α can be obtained; consequently the corrected values of c, α, r_n, r_p and φ will be obtained where φ is the angle between the lines E'O and OA in Fig. 2.

With these corrected values of r_n and r_p the real value of θ is calculated and the indices of the atomic plane by whose reflection the spectrum line is caused is determined and the orientation of the crystal can be determined exactly.

According to the principle stated above, the actual corrections were made with the data represented in Table I, and they are represented in Tables II, III and IV. In these tables the data which are reproduced from Table I are denoted with dashes.

Table II

No. of spots		2θ in degree	$2\delta\theta'$	$2\theta_n$ $(2\theta \mp 2\delta\theta')$	mean value of $2\theta_n$	$2\theta'_n$	$\tan 2\theta_n - \tan 2\theta'_n$ in 10^{-4}	φ' in degree	x in 10^{-2}	y in 10^{-4}
1 (100)	α	25° 55.′5	−5°12.′2	31°7.′7	31°3.′1	31°39.′5	−145.2	1°	−13.2	+1.8
	β	23° 4.′1	−7°54.′4	30°58.′5						
2 (321)	α	49° 39′	+6°18.′6	43°20.′4	43°25.′7	43°2.′6	126.6	217°	+54.2	−0.6
	β	43° 58′	+ 27.′0	43°31′						
3 (100)[2]	α	36° 57′	−7°57.′3	44°54.′3	44°54.′3	44°30.′6	136.4	196°	+13.1	−7.0
4 (321)	α	43° 8.′3	+2° 8.′6	40°59.′7	40°59.′4	40°29.′0	145.5	75°	−24.1	+5.1
	β	43° 58.′2	+2°59.′1	40°59.′1						

Table II (Cont.)

No. of spots		2ϑ in degree	$2d\vartheta'$	$2\vartheta_n$ $(2\vartheta \mp 2d\vartheta')$	mean value of $2\vartheta_n$	$2\vartheta'_n$	$\tan 2\vartheta_n - \tan 2\vartheta'_n$ in 10^{-4}	φ' in degree	x in 10^{-2}	y in 10^{-4}
5 (211)	α	31° 55′	−4°19.′6	36°14.′6	36°18.′9	35°48.′3	136.2	164°	−45.6	+5.3
	β	28° 22.′7	−8° 0.′6	36°23.′3						
6 (110)	α	18° 15.′3	0	18°15.′3	18°15.′3	17°56.′2	62.5	133° 30′	−143.0	−35.1

$$\frac{c_0}{a} = 144.6 \times 10^{-4}, \quad \frac{\Delta c}{a} = -3.2 \times 10^{-4}, \quad \frac{c}{a} = 141.4 \times 10^{-4}$$

$$\alpha_0 = 188°32' \qquad \Delta\alpha = 1°53' \qquad \alpha = 190°25'$$

Table III

	$c = 0.417$ mm.	$\alpha = 190°25'$	
No. of spots	$\varphi' - \alpha$ in degree	$dc = c \cos(\varphi' - \alpha)$ in mm.	φ (corrected) in degree
1	−189° 25′	−0.41	49′
2	+26° 35′	+0.37	216° 37′
3	+ 5° 35′	+0.42	195° 55′
4	−15° 25′	+0.40	175° 15′
5	−26° 25′	+0.37	164° 29′
6	−56° 55′	+0.23	135° 32′

Table IV

No. of spots		r' (obs.) in mm.	dc in mm.	r (corrected) in mm.	2θ (calc.) in degree		$d\theta$	θ (calc.)	Grating constant d in 10^{-8}cm.			Davey's date		Indices of the atomic plane
												d in $\frac{-8}{10}$ cm	Indices of atomic plane	
	r_n	18·19		17·78	θ_n	31° 47′								
									dα′	1·59	(1·56)	1·58	(100)	
									dα	1·58	(1·54)	1·58	(100)	
1	r_{p1}	16·39		15·98	θ_{p1}	28°26·′7	2°38′	12°54·′35	dβ	1·41	(1·38)			$(100)_\alpha$
			−0·41						dγ	1·39	(1·35)			
									dα′	1·79	(1·73)			
									dα	1·78	(1·72)			
	r_{p2}	15·46		15·05	θ_{p2}	27°1·′8	4°2·′9	11°29·′45	dβ	1·58	(1·53)	(1·58)	(100)	$(100)_\beta$
									dγ	1·56	(1·51)			
	r_n	27·55		27·92	θ_n	43°25·′4								
									dα	0·848	(0·853)	0·843	(321)	
									dα	0·843	(0·848)	0·843	(321)	
	r_{p1}	30·76		31·13	θ_{p1}	46°32·′4	3° 7′	24°49·′7	dβ	0·752	(0·756)			$(321)_\alpha$
2			+0·37						dγ	0·738	(0·742)			
									dα′	0·953	(0·964)			
									dα	0·947	(0·958)			
	r_{p2}	27·77		28·14	θ_{p2}	43°28·′9	13·′5	21°56·′2	dβ	0·845	(0·855)	0·843	(321)	$(321)_\beta$
									dγ	0·829	(0·839)			
	r_n	29·00		29·42	θ_n	44°55·′4								
									dα′	1·12	(1·14)	1·12	$(110)^2$	
3									dα	1·12	(1·13)	1·12	$(110)^2$	$(110)^2_\alpha$
	r_p	25·21	+0·42	25·63	θ_p	40° 59′	3°56·′4	18°31·′3	dβ	0·994	(1·01)			
									dγ	0·976	(0·989)			

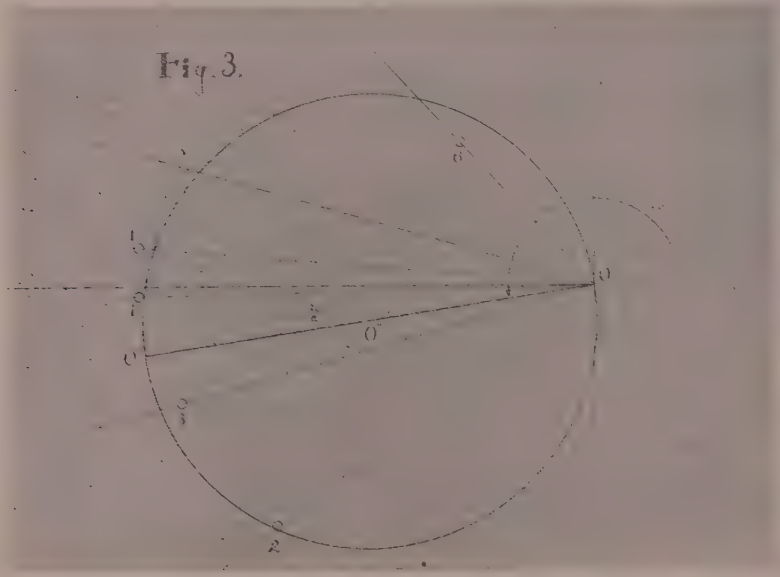
Table IV (cont.)

No. of spots		r' (obs.) in mm.	dc in mm.	r (corrected) in mm.	2θ (calc.) in degree		$d\theta$	θ (calc.)	Grating constant d in 10^{-8}cm.			Davy's date d in 10^{-8} cm	Indices of atomic plane	Indices of the atomic plane
	r_n	25·18		25·58	θ_n	4°0′51·″8								
									$d_{\alpha'}$	0·970	(0·980)			
									d_α	0·965	(0·974)			
	r_{p1}	26·15		26·55	θ_{p1}	41°59·″3	1°3·″5	21°31·″4	d_β	0·860	(0·862)			$(321)_\gamma$
4			+0·40						d_γ	0·845	(0·853)	0·843	(321)	
									$d_{\alpha'}$	0·953	(0·961)			
									d_α	0·947	(0·956)			
	r_{p2}	26·54		26·94	θ_{p2}	42°24·″2	1°28·″4	21°56·″3	d_β	0·845	(0·852)	0·843	(321)	$(321)_\beta$
									d_γ	0·829	(0·837)			
	r_n	21·28		21·65	θ_n	36°16·″5								
									$d_{\alpha'}$	1·29	(1·31)	1·29	(211)	
									d_α	1·28	(1·31)	1·29	(211)	
	r_{p1}	19·63		20·00	θ_{p1}	34°8·″2	2°8·″3	15°50·″95	d_β	1·15	(1·16)			$(211)_\alpha$
5			+0·37						d_γ	1·12	(1·14)			
									$d_{\alpha'}$	1·45	(1·48)			
									d_α	1·45	(1·47)			
	r_{p2}	18·29		18·66	θ_{p2}	32°18′·6	3°57·″9	14°10·″35	d_β	1·29	(1·31)	1·29	(211)	$(211)_\beta$
									d_γ	1·23	(1·29)			
	r_n	9·55		9·78	θ_n	18°20·″5								
									$d_{\alpha'}$	2·23	(2·28)	2·23	(110)	
6									d_α	2·22	(2·27)	2·23	(110)	
	r_p	9·55	+0·23	9·78	θ_p	18°20·″5	0	9°10·″25	d_β	1·98	(2·02)			$(110)_\alpha$
									d_γ	1·94	(1·99)			

Now, assuming that the spectrum lines, 1, 2, 3, etc., are caused respectively by the crystal plane shown in the last column of Table I, the corresponding values of the glancing angle θ were calculated. When the spot contains two spectrum lines the mean of two values of θ obtained with these two lines is tabulated. For the values of the grating constants the writer has taken the Davey[1] data and for the wave lengths the same values as in the case of Table I were used.[2]

The difference between $\tan 2\theta_0$ and $\tan 2\theta_0'$ are calculated and given in the 8th column of Table II, and by using the observed values of φ' in the 9th column the writer plotted the relation expressed by equation (5) as shown in Fig. 3. Keeping the distribution of the plotted spots in mind the circle OO′ and the diameter OO″O′ which passed through the origin were drawn. The length of this diameter and its vectorial angle are denoted by $\frac{c_0}{a}$ and α_0 respectively. With these values of $\frac{c_0}{a}$ and α_0 the writer calculated the values of $x=\cos(\varphi'-\alpha_0)$ and $x=\tan(\varphi'-\alpha_0)$, and plotted the relation expressed by equation (10) in Fig. 4 for the spots 1, 2, 3, etc. These data are represented in Table II, where all symbols represent the same meaning as in the case of

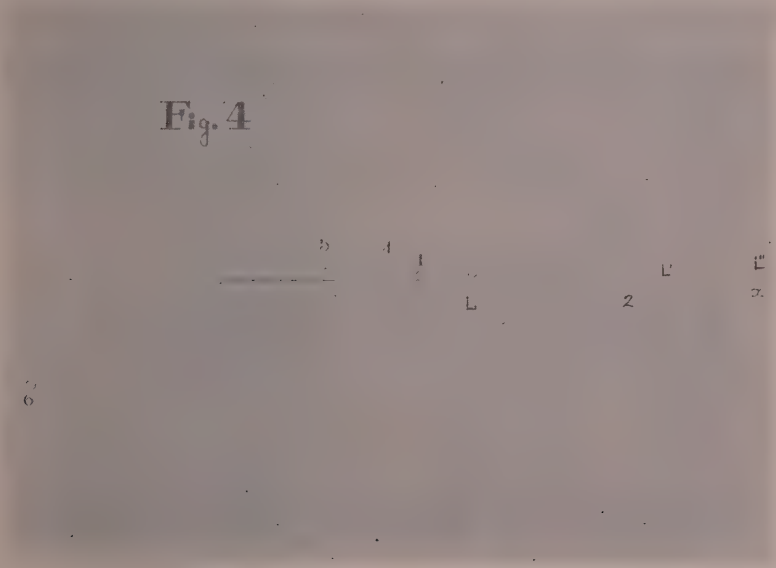
Fig. 3.



1 & 2, loc. cit.

Table I, and the values of $2d\theta'$ and $2\theta_n'$ in the 4th and 7th columns respectively are reproduced from Table I.

Fig. 4



In Figs. 3 and 4, the spots 1, 2, 3, etc., refer respectively to those represented in Fig. 3, Plate I, and Tables I and II. In Fig. 3 one division corresponds to 10^{-4} and in Fig. 4 one division of the abscissa and one of the ordinate correspond to 10^{-2} and 10^{-4} respectively. In Fig.4 it can easily be seen that the spots lying outside of the circle OO′ in Fig. 3, are in the 1st and 2nd quadrants, and those lying inside the circle in Fig. 3 are in the 3rd and 4th quadrants. From the position and the inclination of the line LL′ drawn in Fig. 4 the values of $-\frac{\Delta c}{a}=OL$ and $\frac{c_0}{a}(\Delta\alpha)=\tan L''L'x$ were obtained, and by getting the values of $\Delta\alpha$ by calculation, the corrected values of c and α are obtained and shown at the bottom of Table II.

The corrected values of $dc=\frac{c}{a}\cos(\varphi'-\alpha)$ and φ were calculated for all spots and are shown in Table III.

With the values of dc thus corrected the writer obtained the corrected values of r_n and r_p and calculated the corrected values of θ for all spectrum lines obtained. With these values of the glancing angle, the grating constants of the various atomic planes were recalculated and

compared with the Davey data in Table IV. In this table all symbols have the same meaning as those in Table I. For comparison the uncorrected grating constants represented in Table I are reproduced in brackets in the 12th column of the same table.

It will be easily seen from the table that the corrected values of d are in better agreement with those obtained by Davey than those uncorrected. Thus we can determine more accurately the indices of the atomic plane by whose reflection the spectrum line is caused, and also we can determine more accurately the orientation of the crystallographic axes of the crystal.

The Method using Crossed Wires

The method of taking the photograph was improved by the writer so as to obtain the exact position of the centre of the intense nucleus of the central spot which is impressed by the direct beam of the X-rays.

Before the setting of the photographic plate, crossed metallic wires of high atomic number were so stretched and fixed, between the specimen and the position where the photographic plate would be placed, that the line connecting the centre of the slit and the crossed point of the wires became perpendicular to the plane of the crossed wires. Then the position of the X-ray tube was so adjusted as to project the image of the centre of the focus at the crossed point, and then the photographic plate was placed parallel to the plane containing the crossed wires behind it. For the crossed wires an unannealed tungsten wire was used, the diameter of which was about 0.07 mm. In adjusting the position of the X-ray tube a fluorescent screen was used at first, and then fine adjustment was made by taking a pin hole photograph of the focus of the target as shown in Fig. 9 of Plate II.

With this apparatus a Laue diffraction pattern of thin tungsten wire composed of a single crystal was taken and is represented in Fig. 10 of Plate II. In this photograph we can see the shadow of the crossed wires in the region outside the intense central spot. Though the intensity of the central spot is too strong and the position of the crossed point can hardly be detected in the central part of the central spot, its position is obtained exactly and easily by elongating the detectable part of the shadow-lines.

By setting initially the axis of the specimen parallel to one of the crossed wires, the direction of the axis of the specimen can be obtained

exactly on the photograph.

The correction before mentioned, in addition to the method mentioned above, will give us a more exact position of the centre of the direct image of the focus of the target.

In conclusion, the writer wishes to express his sincere thanks to Professor U. Yoshida, of Kyoto Imperial University for his invaluable suggestions.

Research Laboratory,
Tokyo Electric Company,
Kawasaki, Kanagawa-Ken,
Japan.

Plate I

Fig. 1

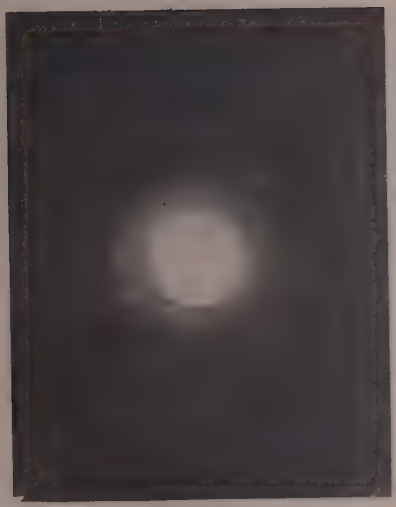


Fig. 2

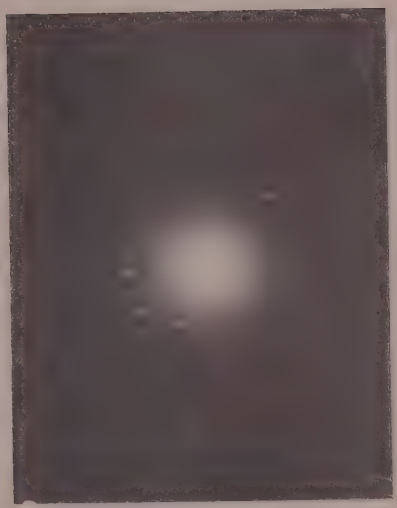


Fig. 3

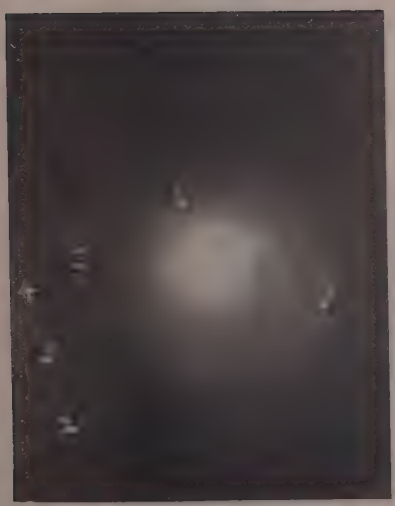


Fig. 4

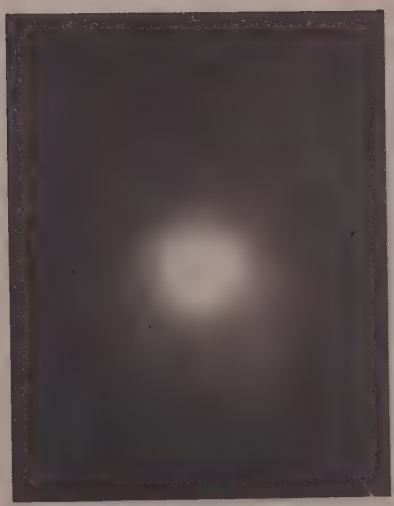


Plate II

Fig. 5

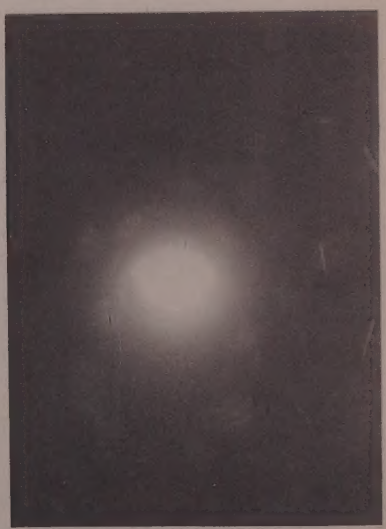
Fig. 6

Fig. 7

Fig. 8

Fig. 9

Fig. 10



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